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NOBLE GASES IN THE MOON

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University of Missouri
Department of Chemistry
Rolla, Missouri 65401

O. K. Manuel
B. Srinivasan
E. W. Hennecke
D. E. Sinclair

NOBLE GASES IN THE MOON

The present report represents a summary of the work performed during the first year of an investigation supported by the National Aeronautics and Space Administration, NASA-NGR-26-003-057. The results of our studies are contained in three Appendices, and we will briefly summarize each following an introduction to the purposes of this investigation.

I. Introduction

Our laboratories have been actively involved for several years in the effort to decipher the origin of isotopic anomalies of noble gases in terrestrial and meteoritic samples. The problem is not simply an academic one; the efforts of many investigators have shown that the noble gas isotopes contain one of the most comprehensive records of geologic events. Due to the great depletion of noble gases in planetary material and to the multitude of nuclear processes which have altered the isotopic composition of these elements over geologic time, in many samples there is little or no record of the primitive noble gases originally incorporated into the sample. Further, recent studies have shown that fractionation of the isotopes in transport processes have severely altered the composition of noble gases in some samples.

In order to decipher the record of events (radioactive decay, nuclear reactions and fractionation processes) which have altered the isotopic composition of noble gases in planetary material, information on the isotopic composition of the primitive noble gases originally associated with planetary

material is needed. According to our present understanding of the early solar system, the primitive noble gases are expected to have been identical in isotopic composition to the noble gases presently in the sun, except for possible alterations in the latter from solar nuclear reactions. The return of fine-grained soil from the lunar surface, where gases were implanted by the solar wind, offers man an opportunity to obtain reliable information on the composition of noble gases present when planetary material formed.

The results of our analyses of solar type noble gases are compared with the isotopic composition of noble gases in meteorites and in terrestrial samples in order to obtain a better understanding of the noble gas record of events in planetary material.

2. Noble Gases in Lunar Fines (15601, 64)

In our first analysis of noble gases in lunar material, the gases were extracted from soil (15601, 64) by heating the sample to successively higher temperatures in 100° intervals from 100°C to 1500°C. Differences in the isotopic composition of He, Ne, Ar and Kr released at different extraction temperatures could be accounted for by a combination of spallation products, isotopic fractionation and radiogenic ^{40}Ar . The fractionation pattern seen across the isotopes of the individual noble gases was paralleled by fractionation of the abundances of the different noble gases.

The isotopic composition of Ne, Ar, Kr and Xe implanted in this sample by the solar wind is enriched in the light-weight isotopes relative to the composition of these elements in typical carbonaceous chondrites or in the atmosphere. Relative to fractionated atmospheric xenon, the xenon

implanted in this sample appeared to be enriched in ^{134}Xe and ^{136}Xe but depleted in ^{129}Xe . The xenon in average carbonaceous chondrites deviates from fractionated atmospheric xenon in a similar manner, and our results indicate that the xenon implanted in 15061, 64 resembles fractionated AVCC-xenon more closely than it resembles fractionated atmospheric xenon.

The results of this study are discussed in more detail in the manuscript in Appendix I.

3. Noble Gases in Lunar Fines (15271, 65)

The abundance and isotopic composition of noble gases in lunar soil (15271, 65) were measured at successively higher temperatures from 600°C to 1600°C in 200° intervals. This sample contained more solar-wind implanted gases than did soil (15601, 64) and also more cosmogenic noble gases.

The results of this study are compared with the isotopic composition of noble gases in terrestrial and meteoritic samples to look for evidence of nuclear processes. It was noted that the $^{38}\text{Ar}/^{36}\text{Ar}$ ratio in solar-wind implanted argon in both soil samples is only slightly smaller than the $^{38}\text{Ar}/^{36}\text{Ar}$ ratio in atmospheric argon. This observation suggests the possibility that nuclear processes may have altered the $^{38}\text{Ar}/^{36}\text{Ar}$ ratio in terrestrial or solar argon. Otherwise, the approximate equality of solar and atmospheric $^{38}\text{Ar}/^{36}\text{Ar}$ ratios seem inconsistent with the following observations:

- a) Solar and atmospheric neon are related by fractionation;

- b) The atmosphere is strongly depleted in the lighter-weight noble gases across Ne, Ar and Kr relative to solar noble gases; and
- c) The isotopic composition of He, Ne and Ar in carbonaceous chondrites and in lunar fines display a common fractionation.

The isotopic composition of solar-wind implanted krypton in soil 15271,65 is slightly depleted in the light isotopes relative to atmospheric krypton, and the krypton released at low extraction temperatures shows an additional depletion of the light-weight isotopes in the manner expected from fractionation.

The isotopic composition of xenon in soil 15271,65 shows only small isotopic variations that could be attributed to fractionation. The spallation yields of xenon isotopes in both 15271,65 and 15601,64 appear to have $^{124}\text{Xe}:^{126}\text{Xe} \approx 0.53$, which is below the $^{124}\text{Xe}:^{126}\text{Xe}$ yields reported by Marti and Lugmair for the highly cosmogenic xenon in 12021,61. The solar xenon in 15271,65 appears to have excess ^{136}Xe and ^{134}Xe relative to fractionated atmospheric xenon. In two temperature fractions, the excess ^{136}Xe and ^{134}Xe are lower than we have observed in any other temperature fraction of lunar soils, but still distinctly above fractionated atmospheric xenon.

The data and figures showing our results for soil 15271,65 are presented in Appendix II.

4. Xenon Released from Carbonaceous Chondrites Near 800°-1000°C

The origin of excess ^{136}Xe and excess ^{134}Xe in the xenon released from carbonaceous chondrites near 800°-1000°C has been considered by several investigators of meteoritic xenon. Recently, we noted that the data from several different laboratories seemed to show a release of excess ^{124}Xe and excess ^{126}Xe in the same temperature fractions which showed excess ^{136}Xe and excess ^{134}Xe . This correlation seems difficult to reconcile with any of the hypothesis that have been advanced to account for the excess ^{136}Xe and excess ^{134}Xe in carbonaceous chondrites. We are confident that the correlation is real in carbonaceous chondrites, but we see no evidence of this behavior in xenon from lunar samples.

The implications of this correlation are discussed in a manuscript in Appendix III.

APPENDIX I

A Comparison of Noble Gases Released from Lunar
Fines (# 15601, 64) with Noble Gases in Meteorites and in the Earth

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A COMPARISON OF NOBLE GASES RELEASED FROM LUNAR FINES
(# 15601.64) WITH NOBLE GASES IN METEORITES AND IN THE EARTH

B. SRINIVASAN*, E. W. HENNECKE, D. E. SINCLAIR and O. K. MANUEL

Division of Earth and Planetary Science

Department of Chemistry

University of Missouri

Rolla, Missouri 65401

*Present address: Department of Physics

University of California

Berkeley, California 94720

Abstract—The abundance and isotopic composition of helium, neon, argon, krypton and xenon which had been released by stepwise heating of lunar soil (15601.64) were measured mass spectrometrically. The extraction temperatures ranged from 100°C to 1500°C in 100° intervals. The bulk of the noble gases were released between 700°C and 1500°C, and the release pattern over this temperature range showed a preferential release of the lighter weight noble gases at low extraction temperatures. This release pattern was also observed across the isotopes of the individual noble gases. Very little gas was released below 700°C, but the gas release pattern from the very low extraction temperatures was unlike the general release trend, showing an enrichment of the heavier noble gases. This anomalous release pattern at low extraction temperatures ($T \leq 700^\circ\text{C}$) was also observed across the isotopes of individual noble gases and is attributed to one, or a combination, of two effects: (a) adsorption of atmospheric gases on the lunar soil during its residence on earth, or (b) selective release of the lighter weight noble gases from the low-temperature sites of lunar soil during its residence on the lunar surface. Isotopic release patterns of noble gases from lunar soil are compared with atmospheric and meteoritic noble gases. Our results indicate that major differences between the isotopic composition of He, Ne, Ar and Kr in terrestrial, meteoritic and lunar samples can be accounted for by a combination of mass fractionation and in situ nuclear processes in solid planetary material. However, it is evident that additional processes, perhaps nuclear reactions in the sun, have altered the isotopic composition of xenon. Relative to fractionated atmospheric xenon, the spallation-free xenon in lunar fines appears to be enriched in the heavy isotopes, ^{134}Xe and ^{136}Xe , and depleted in ^{124}Xe and ^{129}Xe .

INTRODUCTION

The noble gases trapped in solid planetary material contain a record of many geologic events. Due to their chemical inertness and volatile nature, these elements were almost completely lost from other chemical elements of the planetary system when solid bodies formed. Studies of the abundance of noble gases in meteorites and in the earth show that the heavier noble gases have been preferentially retained, and the fractionation pattern across the noble gases provides a record of the conditions which prevailed when these gases separated from more condensable matter in the early history of the solar system (SUESS, 1949; BROWN, 1949; CANALAS et al., 1968).

Over the history of the solar system, induced nuclear reactions and natural radioactive decay of the more abundant elements have produced many changes in the isotopic composition of the residual noble gases in planetary material. The present isotopic composition of noble gases shows the cumulative effect of many nuclear processes, and in some meteorites almost the entire inventory of noble gases is the result of nuclear reactions or radioactive decay. Trapped noble gas isotopes which could not be accounted for by nuclear processes in a meteorite were discovered by GERLING and LEVSKII (1956). Subsequent studies on noble gases trapped in meteorites showed that the relative abundance of the noble gases followed the same general fractionation pattern observed for atmospheric noble gases, and the possibility of isotopic anomalies resulting from mass fractionation were noted (REYNOLDS, 1960a; ZÄHRINGER

and GENTNER, 1960; KRUMMENACHER et al., 1962; SIGNER and SUESS, 1963). Further studies showed that many brecciated-type meteorites contained large amounts of the light weight noble gases (KOENIG et al., 1961; HINTENBERGER et al., 1962) which had been implanted in the dark colored matrix by the solar wind (SUESS et al., 1964; EBERHARDT et al., 1965a,b; HINTENBERGER et al., 1965).

Analyses on the first lunar samples returned to earth (LSPET, 1969) demonstrated that lunar fines and breccias contain large amounts of noble gases from the solar wind. This study of noble gases released from lunar fines (# 15601.64) was undertaken in order to obtain quantitative information on noble gas isotopes which have been implanted by the solar wind. A comparison of the isotopic composition of noble gases in planetary material with the isotopic composition of noble gases presently being implanted by the solar wind is useful in identifying the effects of (a) radioactive decay and induced nuclear reactions which have occurred in planetary bodies, (b) mass fractionation, and (c) solar nuclear reactions which have occurred since the separation of planetary material from the elements in the sun.

EXPERIMENTAL PROCEDURE

An aliquot of soil sample 15601.64 weighing 0.9861 g was used in this study. This soil consists of fine < 1 mm. The sample was wrapped in aluminum foil and mounted in a quartz side-arm chamber of an extraction bottle. After the pressure had been reduced to 10^{-8} torr, a blank Al foil in another quartz side arm chamber was heated stepwise from 100°C

through 600°C by resistance heating. Following analyses of these blanks, the sample was heated stepwise through the same temperatures. Higher extraction temperatures were achieved by dropping the sample into a previously degassed molybdenum crucible heated by R. F. induction. Each extraction temperature was maintained for thirty minutes.

The gases released from the sample were cleaned on Cu-CuO at 550° and on Ti at 850°C, then transferred into a secondary clean-up system by adsorption on charcoal cooled with liquid nitrogen. After 15 minutes the secondary clean-up system was isolated and the gases cleaned on a fresh Ti surface at 850°C. The gases were admitted into a REYNOLDS' (1956) high sensitivity mass spectrometer as four fractions: helium and neon, argon, krypton, and finally xenon.

The procedures for gas analyses and the data reduction have been described by HENNECKE and MANUEL (1971), except for the following modification. In order to measure the isotope ratios for extraction temperatures which released large amounts of gas, small aliquots ($\approx 1/100$ and $\approx 1/30$) of the gases in the secondary clean-up system were isolated and analyzed after the regular gas analysis procedure was complete.

The errors reported in the isotopic ratios for noble gases represent one standard deviation (σ) from the least squares line through the observed ratios plotted as a function of time. The ratios reported are calculated from the least squares lines at the point of entry of the gases into the mass spectrometer. Mass discrimination across the heavier noble gases was determined in the air spikes. The discrimination across

the neon isotopes was less than 0.5% and the discrimination across the two helium isotopes was not measured due to the low atmospheric abundance of ^3He . Except for the three lightest noble gases, all of the isotopic ratios have been corrected for mass discrimination. The concentration of each noble gas was determined from its peak height relative to the average peak height observed in two air spikes analyzed before and after the sample analysis.

RESULTS AND DISCUSSION

In comparing the isotopic composition of noble gases extracted from our lunar soil sample with the isotopic composition of noble gases observed in meteorites or in the earth's atmosphere, the following equation for diffusive fractionation is used (ASTON, 1933; KURODA et al., 1971):

$$\log r = [(m_2 - m_1)/(m_2 + m_1)] [\log (V_i/V_f)] \quad (1)$$

This equation refers to a particular noble gas, and r is the enrichment of the heavy isotope of mass m_2 relative to a lighter isotope of mass m_1 which results when the volume of a gas is reduced from some initial volume, V_i , to some final volume, V_f , by diffusive gas loss. Our use of eq. (1) should not be considered as an indication that we believe all isotopic fractionation effects in noble gases are due to diffusive fractionation. We suspect that many different mass dependent processes may account for part of the complex relationship between planetary and solar-implanted

gases in lunar soil. Eq. (1) is used in order to make our comparisons quantitative (and at the same time to indicate our bias for diffusive gas loss as a major effect in the relationship between planetary and solar noble gases).

Changes in the relative abundance of three or more isotopes of a noble gas due to mass fractionation can be calculated from eq. (1). However, one cannot use eq. (1) to distinguish between mass fractionation and nuclear reactions for gases, such as helium and argon, which have less than three non-radiogenic isotopes.

Helium, Neon and Argon

The results of our analyses for helium, neon and argon are shown in Table 1. The isotopic ratios for argon released at 100°C and 200°C and the isotopic ratios for neon released at 200°C have been corrected for signals observed in blanks, but the signals observed from blank analyses were negligible relative to the sample signals for all other data shown in Table 1.

The $^4\text{He}/^3\text{He}$ ratios increase with increasing extraction temperature from 300°C to 700°C, where the bulk of the helium is released. HOHENBERG et al. (1970) previously observed a preferential release of ^3He from lunar soil 10084 at low extraction temperatures, and suggested that the separation was due to mass fractionation. In Table 1 the highest value for $^4\text{He}/^3\text{He}$ occurs at 700°C, and the $^4\text{He}/^3\text{He}$ ratio in the 700° fraction is 24% higher than the $^4\text{He}/^3\text{He}$ ratio released at 300°C. In the stepwise heating of Apollo 11 lunar soils HOHENBERG et al. (1970) and PEPIN et al. (1970b) observed that the $^4\text{He}/^3\text{He}$ ratio for the total helium was higher than the $^4\text{He}/^3\text{He}$ ratios in the helium released at low extraction temperatures by 36% and 30%, respectively. Figure 1 compares the $^4\text{He}/^3\text{He}$ ratios in sample 15601.64 with the amount of ^4He released at each extraction temperature. The ratios, $^4\text{He}/^3\text{He}$, correlate with the amount of ^4He released over the entire range of extraction temperatures. The correlation

below 700°C may be due to isotopic fractionation, and the correlation above 700°C may be due to the release of cosmogenic ^3He at high extraction temperatures. However, it is not possible to unambiguously distinguish between mass fractionation and nuclear processes from the helium results alone.

The neon isotopes released from sample 15601.64 are shown in Figure 2, where the effects of isotopic mass fractionation and spallation reactions can be separated in the manner employed by MANUEL (1967). The approximately vertical lines extrapolate to cosmogenic neon, and the line through atmospheric neon shows the relationship between the neon isotopes which results from fractionation. This approximately horizontal line is defined by eq. (1), and all of the neon isotopes released at extraction temperatures $< 1000^\circ\text{C}$ lie along this line.

From Figure 2 we note that isotopic mass fractionation is responsible for the high $^{20}\text{Ne}/^{22}\text{Ne}$ released at 500°C and that fractionation and spallation effects are both present in the neon released at high extraction temperatures. The neon released at extraction temperatures $\leq 400^\circ\text{C}$ is not as enriched in the light neon isotopes as that released at 500°C , suggesting that fractionation observed in the neon isotopes released from lunar soil may be a mixture of several mass dependent processes. The pattern of neon isotopes released from 15601.64 is very similar to the release pattern of neon isotopes reported from the gas-rich regions of Fayetteville (MANUEL, 1967), where the highest $^{20}\text{Ne}/^{22}\text{Ne}$ ratio was observed at 600°C and the neon isotopes released at lower extraction temperatures fit the fractionation line shown in Figure 2. The neon isotopes released from lunar soil in this study can be explained in terms of isotopic fractionation and spallation products. A combination of these two processes has previously been proposed to account for the neon isotopes

released by stepwise heating of the gas-rich regions of the Fayetteville meteorite (MANUEL, 1967) and Apollo 11 fines (HOHENBERG et al., 1970). Since lunar soil and the gas-rich meteorites have both acquired neon from solar wind implantation, one should not overlook the possible role of other mechanisms which have been proposed to account for variations of the neon isotopes in gas-rich meteorites (PEPIN, 1967, 1968; BLACK and PEPIN, 1969; BLACK, 1969, 1970).

Table 1 shows the maximum release of argon from lunar fines 15601.64 in the 900°-1000° fractions, and the argon released in the three earlier temperature fractions (600°-800°C) shows lower $^{38}\text{Ar}/^{36}\text{Ar}$ ratios than the argon released at 900°-1000°C. This enrichment of the lighter isotopes in temperature fractions preceeding the release of the bulk of the argon parallels the release pattern observed for neon and helium. Thus, it appears that the argon isotopes released by stepwise heating of lunar fines also show evidence of mass dependent fractionation, as has been suggested earlier by HOHENBERG et al. (1970). The $^{38}\text{Ar}/^{36}\text{Ar}$ ratios in argon released at 300°-500°C show less enrichment of the lighter isotope, ^{36}Ar , than does argon in the 600°-800° fractions. This release pattern of argon isotopes from the 300°-500° fractions is similar to the release pattern of neon isotopes from the 200°-400° fractions and the release pattern of helium isotopes from the 200° fraction. Variations of the $^{38}\text{Ar}/^{36}\text{Ar}$ ratios in the high temperature fractions are probably due to both spallation and fractionation, as was observed in neon. However, a three isotope correlation plot cannot be used to distinguish spallation and fractionation effects in the isotopes of argon due to the presence

of ^{40}Ar from ^{40}K decay.

In our analyses, the $^{40}\text{Ar}/^{36}\text{Ar}$ ratios in the different temperature fractions of lunar soil show the greatest variation of any ratio of two isotopes of a single noble gas. This is undoubtedly due to the presence of radiogenic ^{40}Ar in the soil sample. HEYMANN et al. (1970) and HEYMANN and YANIV (1970) have shown that some of the radiogenic ^{40}Ar in lunar fines is surface correlated and suggested that this excess radiogenic ^{40}Ar may be due to secondary implantation of argon from the lunar atmosphere. The lowest $^{40}\text{Ar}/^{36}\text{Ar}$ ratio from our sample is observed in the argon released at 1000°C , the temperature fraction which released the greatest amount of ^{36}Ar . This sets an upper limit of $^{40}\text{Ar}/^{36}\text{Ar} \leq 0.56$ in the solar wind.

Krypton

The content and isotopic composition of krypton released from 15601.64 is shown in Table 2. MARTI (1967) and EUGSTER et al. (1967) suggested that krypton in carbonaceous chondrites (AVCC-Kr) may be related to atmospheric krypton (NIEF, 1960) by isotopic fractionation, and MANUEL (1970) noted that some of the variations in the isotopic composition of krypton released from lunar soil fits the pattern expected from mass fractionation. Isotope data on krypton released in several low temperature fractions are not shown in Table 2. The amount of krypton released at extraction temperatures $100^\circ\text{--}400^\circ\text{C}$ was too low for reliable isotopic analysis. The initial mass spectrometer signal for krypton in the 600° fraction was higher than that observed from any of the lower

temperature fractions but for reasons which are not understood, the "peak heights" decreased very rapidly with time rendering isotopic analysis impossible.

Figure 3 shows a plot of the data from Table 2, with mass fractionation lines passing through atmospheric krypton in the manner described by eq. (1). The isotopic ratios of krypton released from lunar fines at extraction temperatures $< 1100^{\circ}\text{C}$ and the isotopic ratios of krypton from average carbonaceous chondrites (AVCC-Kr) cluster along these fractionation lines. The krypton isotopes released from lunar fines at extraction temperatures $\geq 1100^{\circ}\text{C}$ do not follow the fractionation lines, suggesting the presence of a component of krypton isotopes from nuclear processes. Using the spallation yields reported by MARTI and LUGMAIR (1971) for krypton isotopes in rock 12021, we can subtract the contribution of spallation-produced krypton isotopes in a manner analogous to that shown for subtracting the spallation-produced neon in Figure 2.

The krypton isotope ratios, corrected for spallation, are shown in Figure 4. From Table 2 it can be seen that the maximum release of krypton from our sample occurred at 1000°C . The krypton isotopes released in the 500° , 700° - 900° fractions are enriched in the lighter isotopes in the manner expected from isotopic mass fractionation, and the krypton released at 1500°C resembles AVCC type krypton. Since each of the lighter noble gases show evidence for an enrichment of the light mass

isotopes in the temperature fractions preceeding the maximum release of that gas, we suggest that this enrichment of lighter krypton isotopes in the 500°, 700°-900° fractions results from isotopic mass fractionation. A similar low temperature release pattern of krypton isotopes from lunar soil 10084-48 has been reported earlier by PEPIN et al. (1970), who suggested that a new type of spallation krypton might be released in the low temperature fractions.

Xenon

The abundance and isotopic composition of xenon released from lunar fines 15601.64 are shown in Table 3, together with the isotopic composition of xenon in the atmosphere (NIER, 1950) and in average carbonaceous chondrites (EUGSTER et al., 1967; MARTI, 1967). We have used ^{132}Xe as the reference isotope because this isotope is common to two different fractionation lines which seem to relate atmospheric and AVCC xenon (REYNOLDS, 1960a; KRUMMENACHER et al., 1962; EUGSTER et al., 1967; MARTI, 1967), as shown in Figure 5. Also shown is the relationship between "solar" xenon, as estimated by EBERHARDT et al. (1970) from analysis of xenon in lunar fines 10084-47, and atmospheric xenon. The abundance of the ^{129}Xe isotope is not shown in Figure 5 due to the existence of radiogenic ^{129}Xe in meteorites (REYNOLDS, 1960b) and in the earth (BOULOS and MANUEL, 1971).

The greatest amount of ^{132}Xe is released from lunar fines 15601.64 at 1000°C. The xenon released in the preceeding temperature fraction, 900°C, shows a greater enrichment of the lighter isotopes, $^{128}\text{-}^{130}\text{Xe}$, and

a greater depletion of the heavier isotopes, $^{134},^{136}\text{Xe}$, than does the xenon released at 1000°C . The $^{124},^{126}\text{Xe}$ isotopes are not considered now due to the presence of a sizable contribution of spallation products in the 1000° fraction, as will be discussed later. Since the release pattern of xenon isotopes in the temperature fraction preceeding the maximum release of xenon parallels the release pattern observed for all other noble gases in the temperature fractions prior to the release of the maximum amount of each gas, it appears that isotopic fractionation may be responsible for some differences in the xenon released in our stepwise heating experiment.

Prior to analysis of gases released at 800°C , the spectrometer tube was accidentally exposed to the atmosphere. Subsequent analyses of the lighter noble gases seemed to show no evidence for contamination with atmospheric-type noble gases, and it was thought that analyses of these lighter noble gases from the 800° fraction would act to "scrub" the spectrometer of any heavier atmospheric gases. However, before letting the 800° fraction of xenon into the spectrometer we observed instrument background xenon equivalent to about $1/5$ of the xenon in the 800° fraction. Due to the possibility of atmospheric contamination in the 800° fraction of xenon, we will not include this temperature fraction in our discussion of the isotope release pattern of xenon.

The isotopic composition of xenon released at 400° , 500° and 700°C appear to be enriched in the heavy isotopes. Isotopic ratios for xenon released at 600°C are not shown due to a rapid decrease of the spectrometer signal during analysis of these isotopes. No enrichment of heavy krypton

isotopes was observed in low temperature fractions but the 200° fraction of helium, the 300°-400° fractions of neon, and the 300°-500° fractions of argon are more enriched in the heavy isotopes than are the gases released in the next few higher temperature fractions. The reasons for this enrichment of heavy isotopes in low temperature fractions is not clear, but may result from adsorption of atmospheric type gases on the sample prior to analysis or to the selective loss of the lighter noble gas isotopes (from sites which are degassed at low laboratory extraction temperatures) during the time that the soil resided on the lunar surface.

In Table 4 we have shown the cumulative percent release of each noble gas as the extraction temperatures are increased. For extraction temperatures $\leq 500^{\circ}\text{C}$, the cumulative percent release of Xe and Kr exceeds that of the three lighter weight noble gases. This release pattern may be related to the enrichment of heavy isotopes observed in the low extraction temperatures, but the mechanism involved is not understood. For extraction temperatures $> 600^{\circ}\text{C}$, the gas release pattern clearly demonstrates the preferential release of the lighter noble gases. This is shown in Figure 6, where the cumulative percent yield is plotted against extraction temperature.

The isotopic composition of xenon from Table 3 is shown in Figure 7. Solid lines passing through atmospheric xenon fit the fractionation pattern defined by eq. (1), and dashed fractionation lines are shown through ^{134}Xe and ^{136}Xe in AVCC xenon. The ^{124}Xe and ^{126}Xe isotopes show a large scatter due to the presence of spallation components, and

the ^{126}Xe data are shown as dark points in order to distinguish them from ^{124}Xe and ^{128}Xe . The dashed fractionation line labeled 129' assumes that about 5.7% of atmospheric ^{129}Xe is due to radiogenic ^{129}Xe from the decay of extinct ^{129}I (BOULOS and MANUEL, 1971).

Using the spallation yields reported by MARTI and LUGMAIR (1971), the contribution of spallation products to each xenon isotope has been subtracted in the manner employed for neon and krypton. For this correction we first used deviations of the points, ($^{126}\text{Xe}/^{132}\text{Xe}$, $^{130}\text{Xe}/^{132}\text{Xe}$), from the fractionation line to compute the amount of spallation-produced ^{126}Xe . The contribution of spallation to each xenon isotope was then calculated from its spallation relative to that of ^{126}Xe . The xenon spectrum, corrected for spallation products, is shown in Figure 8.

From the isotope correlations shown in Figure 8 we note that there are only small fractionation effects in xenon released from our sample at extraction temperatures $\geq 900^\circ\text{C}$. The behavior of ^{129}Xe , ^{136}Xe and ^{134}Xe isotopes in the 400° and 500° fractions seems to better fit a mixture of atmospheric and AVCC-type xenon than the dashed fractionation lines. The ^{124}Xe seems depleted relative to fractionated AVCC or atmospheric xenon in all temperature fractions. The apparent depletion of ^{124}Xe may be due to (a) an excess of ^{126}Xe in the solar wind, perhaps due to the $^{127}\text{I}(\gamma, n\beta)^{126}\text{Xe}$ reaction in the sun; (b) a depletion of ^{124}Xe in the solar wind, perhaps due to the $^{124}\text{Xe}(n, \gamma)^{125}\text{Xe}$ reaction in the sun; or (c) a smaller $^{124}\text{Xe} : ^{126}\text{Xe}$ spallation yield in our sample than that in lunar rock 12021.

The heavy xenon isotopes, $^{134}\text{-}^{136}\text{Xe}$ released from our sample seem to better fit mass fractionated AVCC xenon than mass fractionated atmospheric xenon. Our results do not support the view of MARTI (1969) that solar-wind xenon can be related to atmospheric xenon by mass fractionation, nor the view of EBERHARDT et al. (1970) that solar-wind xenon confirms the presence of a fission component in AVCC xenon. To account for the enrichment of ^{136}Xe and ^{134}Xe in our sample above the solid fractionation line through atmospheric xenon by fission of a volatile, superheavy element (ANDERS and HEYMANN, 1969) would require unreasonable high abundances of superheavy elements in the early solar system. KURODA and MANUEL (1970) and SABU and MANUEL (1971) have previously discussed the difficulties encountered in explaining AVCC xenon by the fission hypothesis.

An alternate way of viewing the relationship between solar, AVCC and atmospheric xenon has been presented by KURODA and his associates (KURODA and MANUEL, 1970; MANUEL et al., 1970; KURODA et al., 1971; KURODA, 1971). These authors have used the heaviest isotope, ^{136}Xe , as a reference and compared the other isotopic ratios of xenon with the ratios calculated by assuming that variations in the $^{134}\text{Xe}/^{136}\text{Xe}$ ratio result from fractionation. When the xenon data is examined in this manner one finds that xenon in both average carbonaceous chondrites and lunar soil appears enriched in the light isotopes, $^{124}\text{-}^{132}\text{Xe}$, relative to fractionated atmospheric xenon. After subtracting the contribution of cosmogenic xenon from the excess $^{124}\text{-}^{132}\text{Xe}$, KURODA (1971) has suggested that the remaining

excess ^{130}Xe and ^{132}Xe may be due to neutron capture processes, $^{129}\text{Xe}(n,\gamma)^{130}\text{Xe}$ and $^{131}\text{Xe}(n,\gamma)^{132}\text{Xe}$, in the sun. We have normalized the isotopic ratios of xenon from lunar soil, 15601.64, to ^{136}Xe and find that there remains an excess of ^{128}Xe , ^{130}Xe and ^{131}Xe after correcting for spallation and fractionation effects. Any excess ^{132}Xe or ^{129}Xe is very small and within the error limits of the measurement. The amount of excess ^{131}Xe contains a large uncertainty due to the highly variable spallation yield of this isotope in lunar samples. However, the excess ^{128}Xe and ^{130}Xe , which occurs in approximately 1:1 ratio in all temperature fractions, is outside the error limits of the measurement or the spallation yields assumed. These anomalies may contain quantitative information on nuclear processes which have occurred in the evolution of the sun.

Thus, a comparison of our results on xenon from lunar soil with the results of earlier analyses of xenon in meteorites and in the atmosphere shows that either (a) the xenon in chondrites and the xenon implanted in lunar soil contains an enrichment of ^{134}Xe and ^{136}Xe from some unknown source, or (b) the xenon in chondrites and in lunar fines contains the products of nuclear reactions in the sun, or (c) the xenon in the earth's atmosphere came from a different nucleosynthesis source than the xenon in meteorites and in the solar wind. Since the other elements of lunar, meteoritic, terrestrial and solar material appear to have been derived from a common batch of chemicals, it appears that the earth should have had an original inventory of xenon isotopes similar to that in meteoritic and lunar material. If the observed xenon in meteorites and in lunar fines has not been

appreciably altered by nuclear reactions in the sun, then the possibility that atmospheric xenon is from some extra-solar source (item c above) could be experimentally verified if AVCC or solar-type xenon were found within the earth. Although recent analyses in our laboratory (BOULOS and MANUEL, 1971; 1972) of terrestrial xenon from deep gas wells have shown the decay products of two now extinct nuclides, ^{129}I and ^{244}Pu , the nonradiogenic xenon isotopes in these wells more closely resembled atmospheric xenon than AVCC or solar-type xenon.

CONCLUSIONS

The results of this investigation on the abundance and isotopic composition of noble gases released by stepwise heating of lunar soil, sample 15601.64, lead us to the following conclusions:

(1) The fractional amount of each gas released above 600°C shows preferential release of the light weight noble gases.

(2) Due to fractionation effects the isotopic composition of solar-wind implanted gases cannot be unambiguously identified from analysis of gases in lunar soils. However, we estimate the isotopic composition of the non-cosmogenic noble gases trapped in our sample to be $^4\text{He}:^3\text{He} = 2280$; $^{20}\text{Ne}:^{21}\text{Ne}:^{22}\text{Ne} = 13.0:0.033:1.00$; $^{36}\text{Ar}:^{38}\text{Ar}:^{40}\text{Ar} = 1.00:0.185:\leq 0.558$; $^{78}\text{Kr}:^{80}\text{Kr}:^{82}\text{Kr}:^{83}\text{Kr}:^{84}\text{Kr}:^{86}\text{Kr} = 0.021:0.132:0.665:0.665:3.29:1.00$; $^{124}\text{Xe}:^{126}\text{Xe}:^{128}\text{Xe}:^{129}\text{Xe}:^{130}\text{Xe}:^{131}\text{Xe}:^{132}\text{Xe}:^{134}\text{Xe}:^{136}\text{Xe} = 0.00445:0.00425:0.0824:1.03:0.163:0.817:1.00:0.374:0.305$.

(3) At very low extraction temperatures there appears to be a gas enriched in the heavy isotopes and in the heavy noble gases. This is due to adsorption of atmospheric gases or to selective release of the light isotopes from these low temperature sites during the period the sample was on the moon.

(4) The isotopic composition of the noble gases released from lunar fines at different extraction temperatures shows variations due to mass fractionation. Differences in the isotopic composition of He, Ne, Ar and Kr in meteorites, in the earth's atmosphere and in the solar wind can also be accounted for by mass fractionation and reasonable nuclear reactions. The results of this analysis of noble gases from 15601.64 and recent results from our studies of noble gases in terrestrial samples (BOULOS and MANUEL, 1970; 1972) and meteorites (SRINIVASAN and MANUEL, 1971; HENNECKE and MANUEL, 1971) substantiate the view that mass fractionation is responsible for many of the isotopic anomalies of noble gases in nature.

(5) The xenon isotopes released from our sample cannot be accounted for by spallation plus fractionation of atmospheric xenon. Unreasonably high abundances of superheavy elements would be required to account for the "excess" ^{134}Xe and ^{136}Xe in solar type xenon relative to fractionated atmospheric xenon.

(6) The relationship between atmospheric, solar and AVCC xenon is complex. Additional studies on the isotopic composition of xenon in a wide variety of lunar, meteoritic and terrestrial samples are needed to decipher the xenon record of events in the early history of the solar system.

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Table 1. Helium, neon and argon from stepwise heating of lunar fines (15601.64)

Temp. (°C)	ISOTOPE RATIOS					GAS CONTENT ($\times 10^{-3}$ cc STP/g)		
	$^4\text{He}/^3\text{He}$	$^{20}\text{Ne}/^{22}\text{Ne}$	$^{21}\text{Ne}/^{22}\text{Ne}$	$^{38}\text{Ar}/^{36}\text{Ar}$	$^{40}\text{Ar}/^{36}\text{Ar}$	^4He	^{22}Ne	^{36}Ar
100°	-----	-----	-----	-----	215.8 ± 0.8	1.8	0.20	0.015
200°	1922 ± 7	12.66 ± 0.03	0.0307 ± 0.0004	-----	29.8 ± 0.1	38	0.69	0.60
300°	1838 ± 14	13.49 ± 0.02	0.0329 ± 0.0003	0.185 ± 0.002	28.2 ± 0.2	300	3.2	1.5
400°	1864 ± 23	13.48 ± 0.05	0.0325 ± 0.0012	0.186 ± 0.001	33.1 ± 0.2	400	4.0	3.9
500°	1911 ± 9	13.70 ± 0.02	0.0336 ± 0.0001	0.183 ± 0.001	31.8 ± 0.2	870	7.0	6.4
600°	2273 ± 3	13.47 ± 0.02	0.0337 ± 0.0001	0.181 ± 0.001	26.7 ± 0.1	6,400	60	15
700°	2280 ± 10	13.01 ± 0.02	0.0327 ± 0.0001	0.178 ± 0.003	4.14 ± 0.05	450,000	15,000	5,200
800°	2093 ± 16	12.76 ± 0.03	0.0320 ± 0.0002	0.177 ± 0.001	2.50 ± 0.01	28,000	640	1,400
900°	2215 ± 12	12.80 ± 0.05	0.0337 ± 0.0002	0.185 ± 0.001	0.764 ± 0.001	54,000	2,500	25,000
1000°	2268 ± 5	12.82 ± 0.02	0.0372 ± 0.0001	0.186 ± 0.001	0.558 ± 0.001	34,000	2,700	28,000
1100°	1355 ± 3	11.91 ± 0.01	0.0656 ± 0.0002	0.194 ± 0.001	0.835 ± 0.005	260	260	2,300
1200°	2108 ± 23	12.26 ± 0.01	0.0557 ± 0.0001	0.204 ± 0.002	0.811 ± 0.007	950	270	1,800
1300°	1928 ± 27	12.13 ± 0.01	0.0673 ± 0.0003	0.254 ± 0.002	0.855 ± 0.001	430	84	580
1400°	2208 ± 18	12.41 ± 0.01	0.0461 ± 0.0001	0.196 ± 0.001	0.711 ± 0.001	1,300	290	3,100
1500°	2169 ± 22	12.66 ± 0.08	0.0434 ± 0.0004	0.194 ± 0.001	0.700 ± 0.001	130	65	700
Air	7,700,000	9.81	0.0290	0.187	296	-----	-----	---

Table 2. Krypton released from stepwise heating of lunar fines (15601.64)

Temp. (°C)	^{86}Kr Released $\times 10^{-10}$ cc STP/g	^{78}Kr	^{80}Kr	^{82}Kr	^{83}Kr	^{84}Kr	^{86}Kr
500°	0.33	-----	-----	73 ± 1	70 ± 1	342 ± 4	± 100
700°	2.5	2.35 ± 0.02	13.8 ± 0.1	69.4 ± 0.7	68.7 ± 0.5	336 ± 2	± 100
800°	0.35	-----	12.8 ± 0.2	67.9 ± 0.5	67.2 ± 0.5	331 ± 1	± 100
900°	8.0	2.10 ± 0.01	13.3 ± 0.1	67.8 ± 0.2	67.8 ± 0.3	332 ± 1	± 100
1000°	25	2.10 ± 0.01	13.3 ± 0.1	66.9 ± 0.3	66.6 ± 0.3	328 ± 1	± 100
1100°	11	2.76 ± 0.03	15.1 ± 0.1	69.9 ± 0.2	70.2 ± 0.2	331 ± 1	± 100
1200°	8.2	2.34 ± 0.02	13.7 ± 0.1	66.3 ± 0.3	67.6 ± 0.4	327 ± 1	± 100
1300°	3.7	2.71 ± 0.04	14.5 ± 0.3	69.1 ± 0.5	70.8 ± 0.5	332 ± 2	± 100
1400°	14	2.38 ± 0.03	13.7 ± 0.1	66.8 ± 0.4	67.4 ± 0.3	329 ± 1	± 100
1500°	3.2	2.25 ± 0.02	13.4 ± 0.1	66.0 ± 0.3	66.3 ± 0.3	324 ± 1	± 100
Atmosphere	----	2.02	13.0	66.2	66.0	327	± 100
AVCC	----	1.92	12.6	64.9	65.1	323	± 100

Table 3. Xenon released from stepwise heating of lunar fines (15601.64)

Temp. (°C)	^{132}Xe Released $\times 10^{-10}$ cc STP/g	^{124}Xe	^{126}Xe	^{128}Xe	^{129}Xe	^{130}Xe	^{131}Xe	^{132}Xe	^{134}Xe	^{136}Xe
400°	0.04	-----	-----	-----	99.8 ± 0.08	15.3 ± 0.2	79.4 ± 0.9	≈ 100	39.6 ± 0.6	33.6 ± 0.7
500°	0.03	-----	-----	-----	99.1 ± 1.6	15.6 ± 0.2	79.8 ± 1.0	≈ 100	39.4 ± 0.4	32.0 ± 0.5
700°	0.27	-----	-----	9.35 ± 0.12	101.1 ± 1.3	15.9 ± 0.1	80.9 ± 1.0	≈ 100	38.5 ± 0.5	32.6 ± 0.4
800°	0.18	-----	-----	7.24 ± 0.04	98.2 ± 0.5	15.0 ± 0.1	78.4 ± 0.4	≈ 100	38.4 ± 0.2	32.5 ± 0.2
900°	1.2	0.487 ± 0.008	0.467 ± 0.005	8.48 ± 0.07	104.9 ± 0.5	16.5 ± 0.1	82.2 ± 0.5	≈ 100	37.3 ± 0.2	30.5 ± 0.2
1000°	6.8	0.518 ± 0.003	0.526 ± 0.003	8.35 ± 0.04	103.4 ± 0.8	16.2 ± 0.1	82.4 ± 0.5	≈ 100	37.2 ± 0.2	30.3 ± 0.2
1100°	6.2	1.00 ± 0.01	1.38 ± 0.01	9.71 ± 0.03	103.8 ± 0.4	17.2 ± 0.1	86.0 ± 0.3	≈ 100	36.9 ± 0.1	30.3 ± 0.1
1200°	4.9	0.794 ± 0.009	1.06 ± 0.01	9.14 ± 0.10	103.2 ± 1.1	16.9 ± 0.1	84.3 ± 0.9	≈ 100	37.6 ± 0.4	31.0 ± 0.3
1300°	1.6	0.899 ± 0.007	1.22 ± 0.01	9.42 ± 0.05	103.7 ± 0.5	17.0 ± 0.1	85.4 ± 0.6	≈ 100	37.2 ± 0.2	30.8 ± 0.1
1400°	6.1	0.758 ± 0.006	0.959 ± 0.007	9.05 ± 0.06	104.2 ± 0.5	16.9 ± 0.1	84.3 ± 0.5	≈ 100	37.2 ± 0.2	30.4 ± 0.1
1500°	1.7	0.692 ± 0.004	0.857 ± 0.008	8.92 ± 0.06	103.3 ± 0.7	16.6 ± 0.1	83.7 ± 0.5	≈ 100	37.2 ± 0.3	30.3 ± 0.2
Atmosphere	-----	0.357	0.335	7.14	98.3	15.2	78.8	≈ 100	38.8	33.0
AVCC	-----	0.452	0.406	8.09	-----	16.1	81.5	≈ 100	38.1	32.0

Table 4. Cumulative percent of each noble gas released from stepwise
heating of lunar fines (15601.64)

Temp. (°C)	^4He	^{22}Ne	^{36}Ar	^{84}Kr	^{132}Xe
100°	0.00031	0.00090	0.000021	0.0057	0.0094
200°	0.00070	0.0040	0.00090	0.026	0.085
300°	0.059	0.018	0.0030	0.111	0.157
400°	0.128	0.036	0.0088	0.288	0.294
500°	0.278	0.069	0.019	0.735	0.393
600°	1.388	0.338	0.040	1.783	0.831
700°	79.388	69.521	7.621	5.114	1.763
800°	84.208	72.412	9.684	5.566	2.395
900°	93.618	83.770	46.876	15.840	6.470
1000°	99.488	95.835	87.585	48.363	29.992
1100°	99.534	96.983	90.934	63.179	51.240
1200°	99.698	98.138	93.533	73.670	67.958
1300°	99.772	98.516	94.482	78.474	73.487
1400°	99.993	99.809	98.981	95.893	94.318
1500°	100	100	100	100	100

Figure 1. The release pattern of helium from sample 15601.64.

The ^4He content on the left ordinate is in units of
cc STP/g.

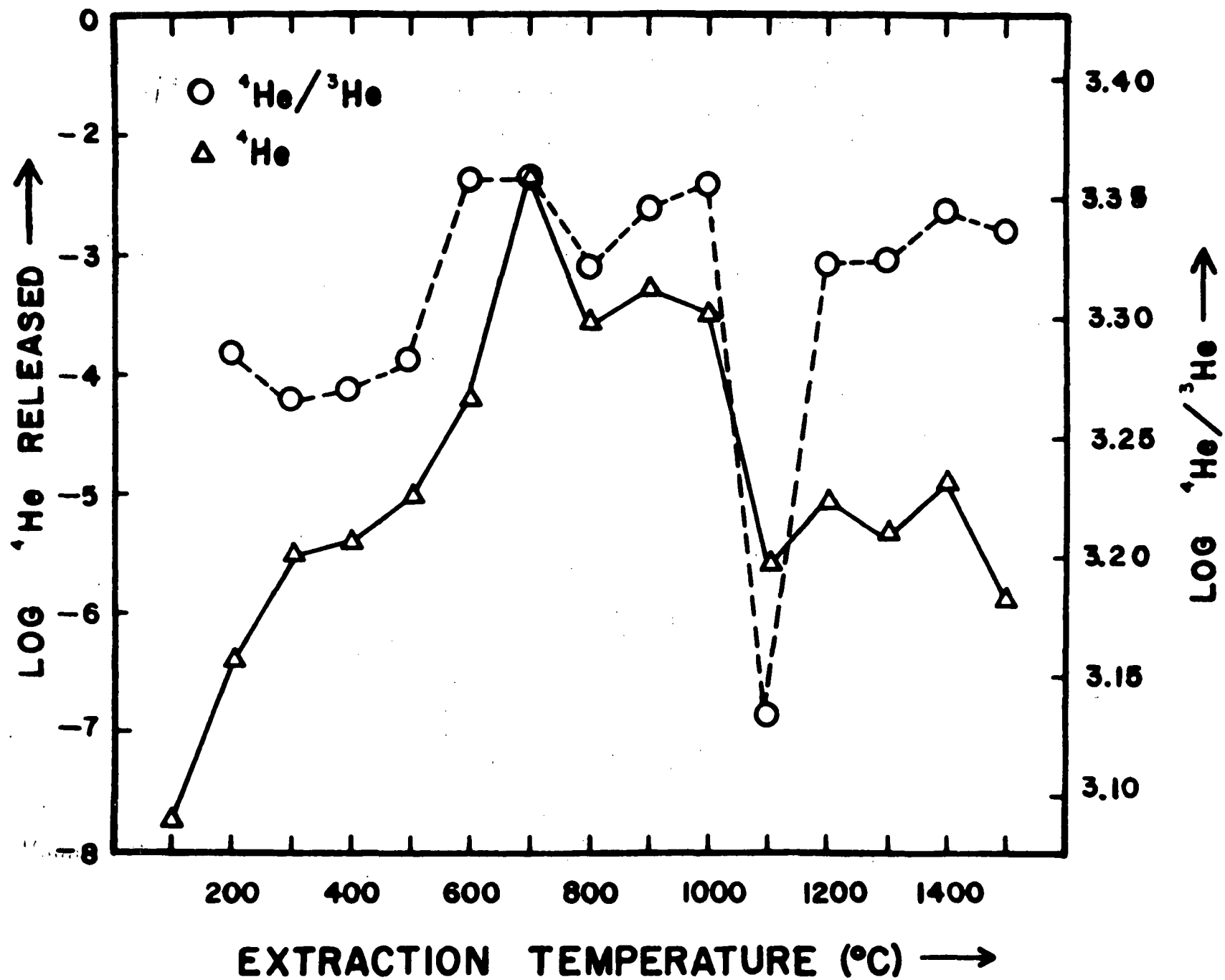


Figure 2. Correlation of the neon isotopes released from sample 15601.64. The triangle represents atmospheric neon and the line through atmospheric neon results from isotopic fractionation. The arrows point toward pure cosmogenic neon, and the intersections of the lines correspond to the isotopic composition of spallation-free neon (MANUEL, 1967).

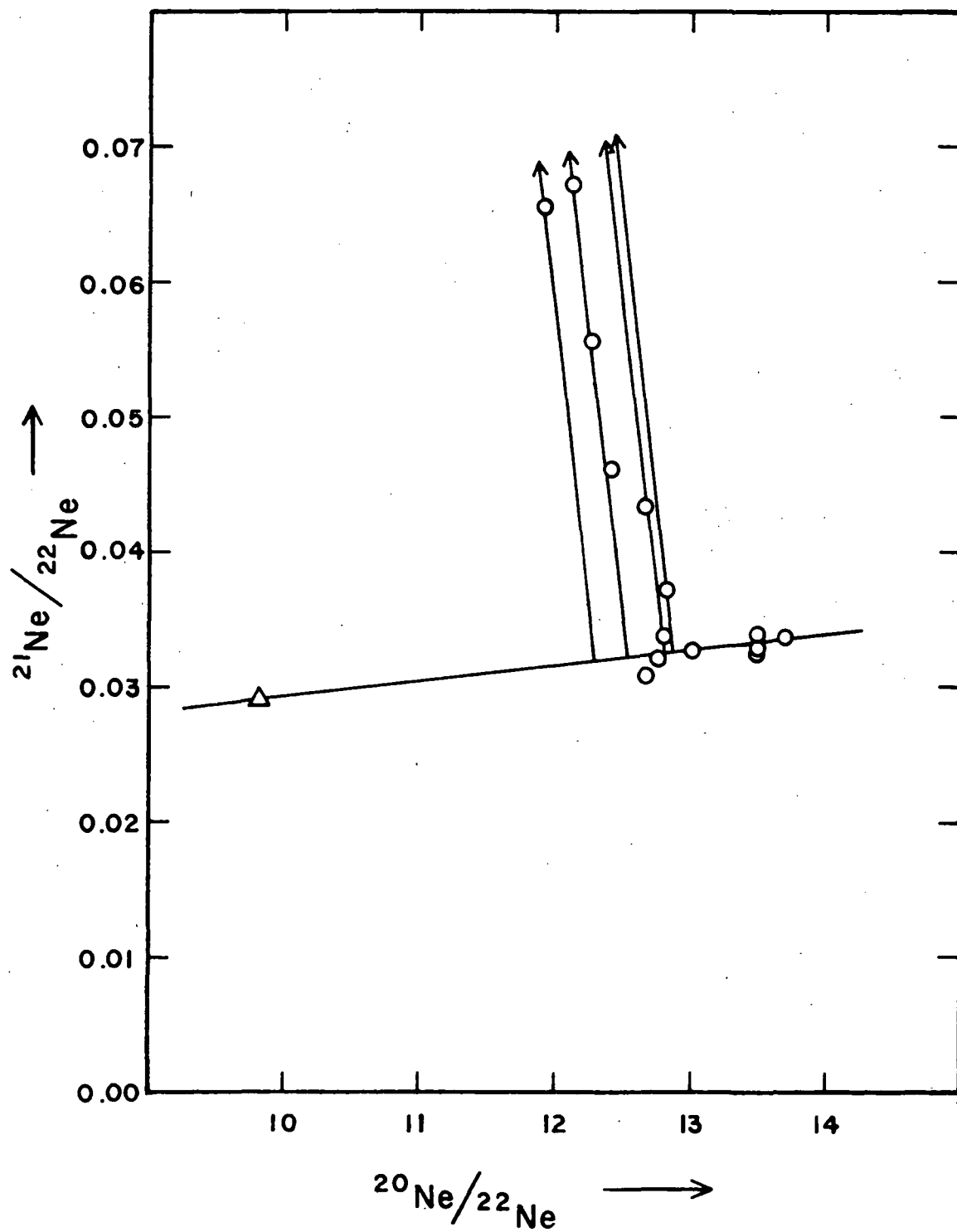


Figure 3. Correlation of the krypton isotopes released from sample 15601.64. The solid lines passing through atmospheric krypton are defined by eq. (1). The scatter of isotope ratios from the correlation lines is due to cosmogenic krypton.

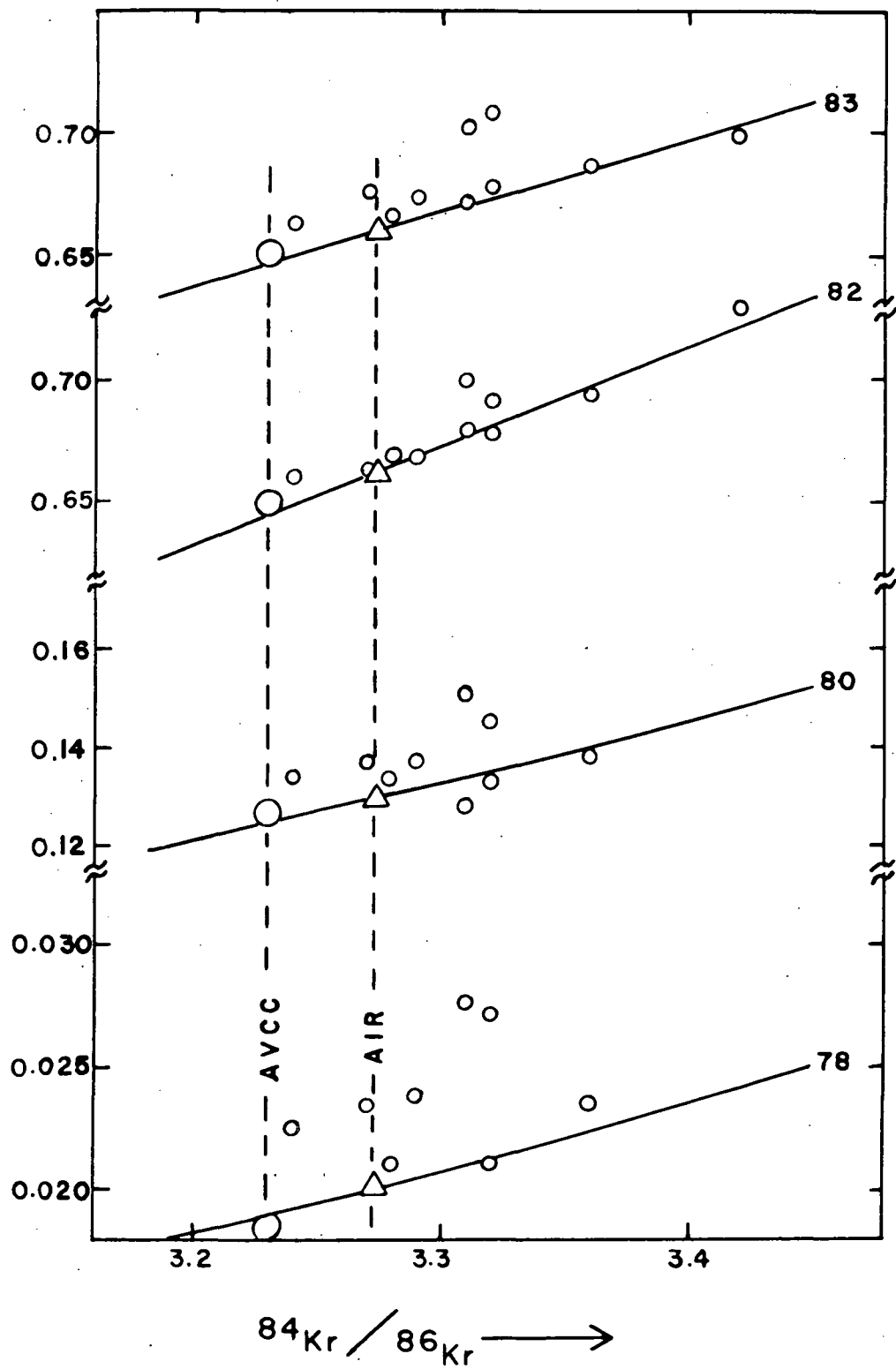


Figure 4. Correlation of the spallation-free krypton isotopes released from 15601.64. The solar-wind implanted krypton isotopes correlate in the manner expected from isotopic fraction, lying on the same correlation line which relates atmospheric and AVCC krypton (MARTI, 1967; EUGSTER et al., 1967).

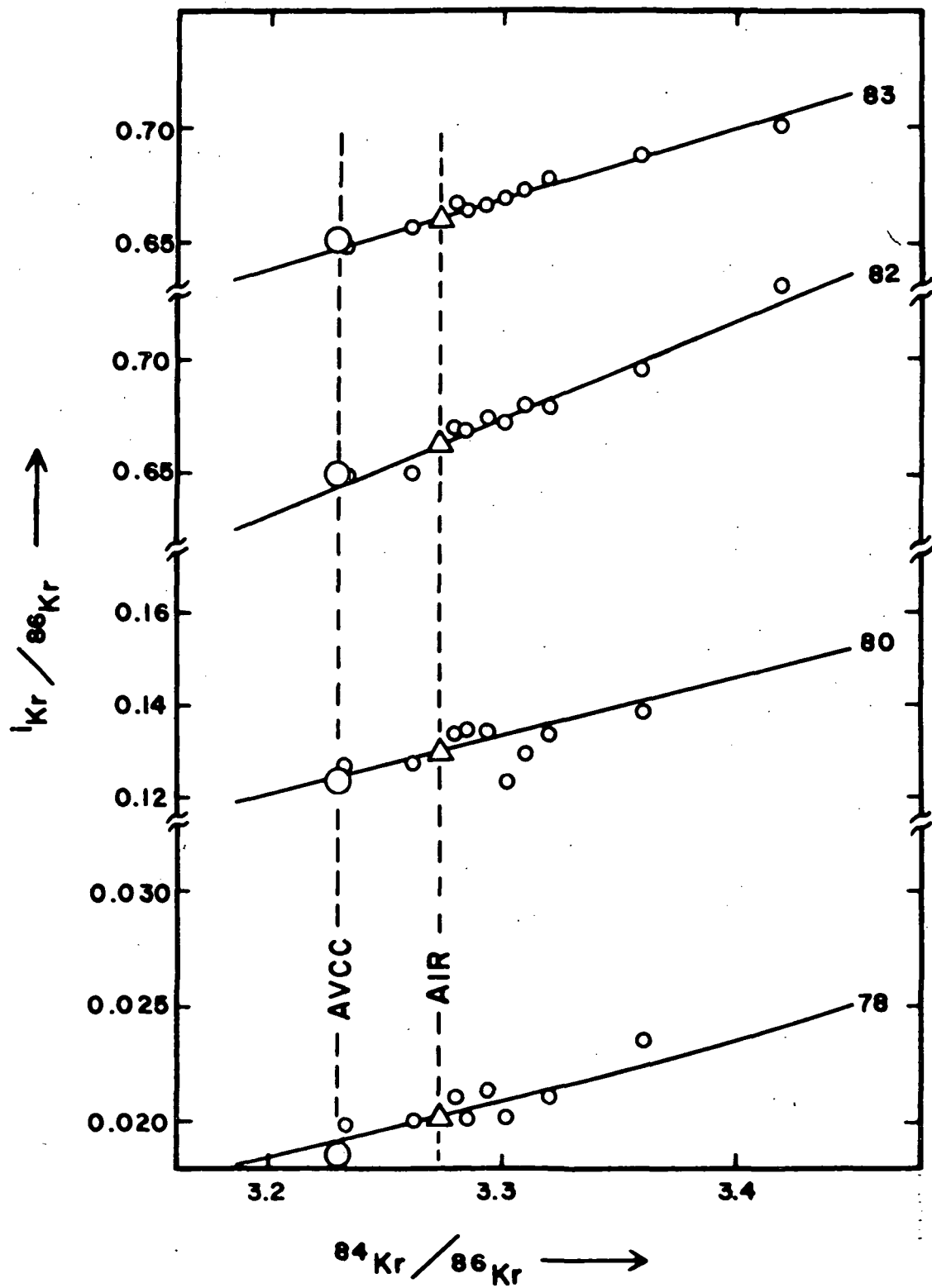


Figure 5. A comparison of the abundances of xenon isotopes in average carbonaceous chondrites (EUGSTER et al., 1967; MARTI, 1967) and in solar-type xenon (EBERHARDT et al., 1971) with the abundance of xenon isotopes in the earth's atmosphere (NIER, 1950). Fractionation across the heavy isotopes $^{132}\text{-}^{136}\text{Xe}$ appears to be less steep than fractionation across the light isotopes $^{124}\text{-}^{132}\text{Xe}$, and the ^{132}Xe isotope is common to both fractionation patterns.

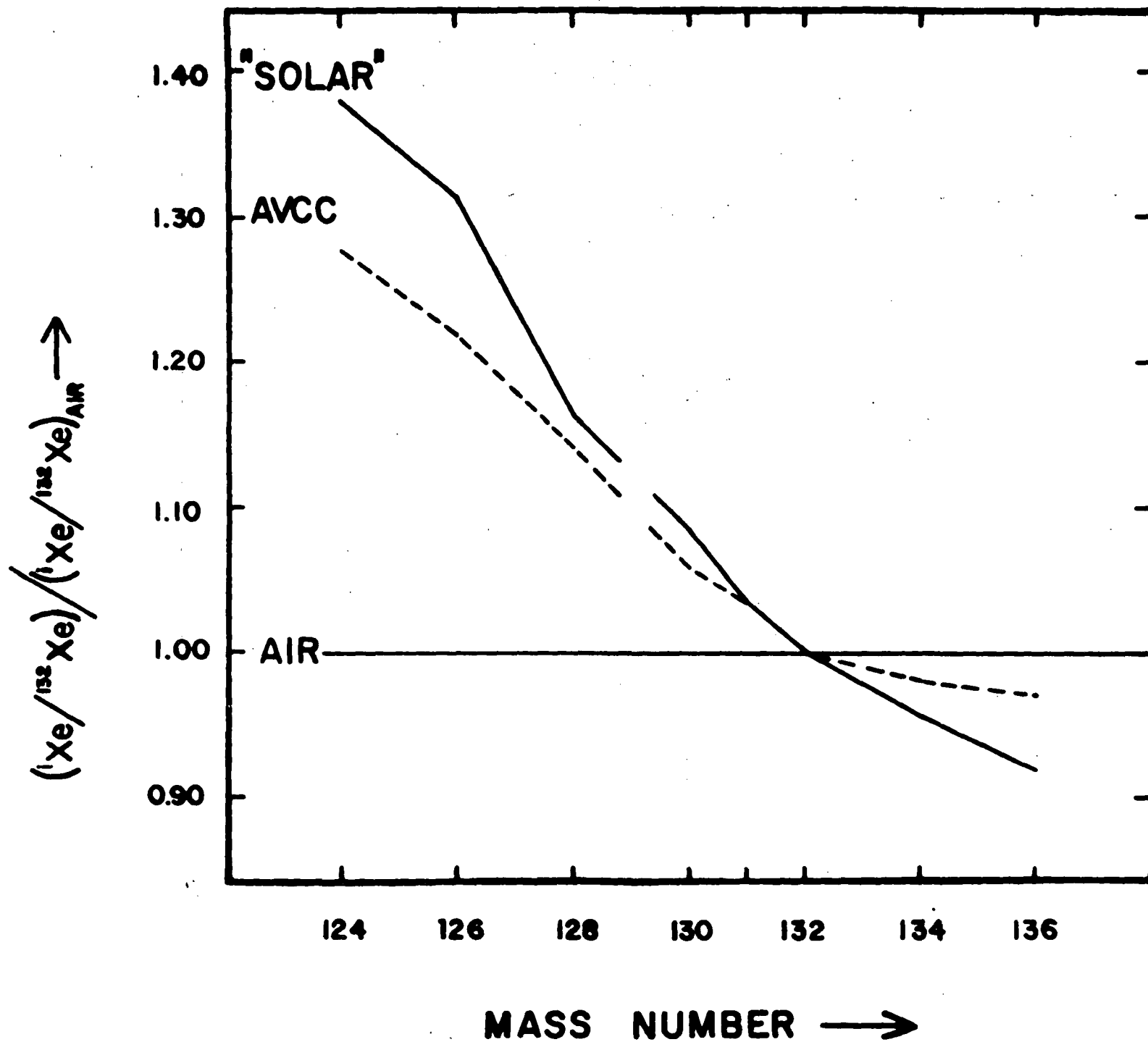
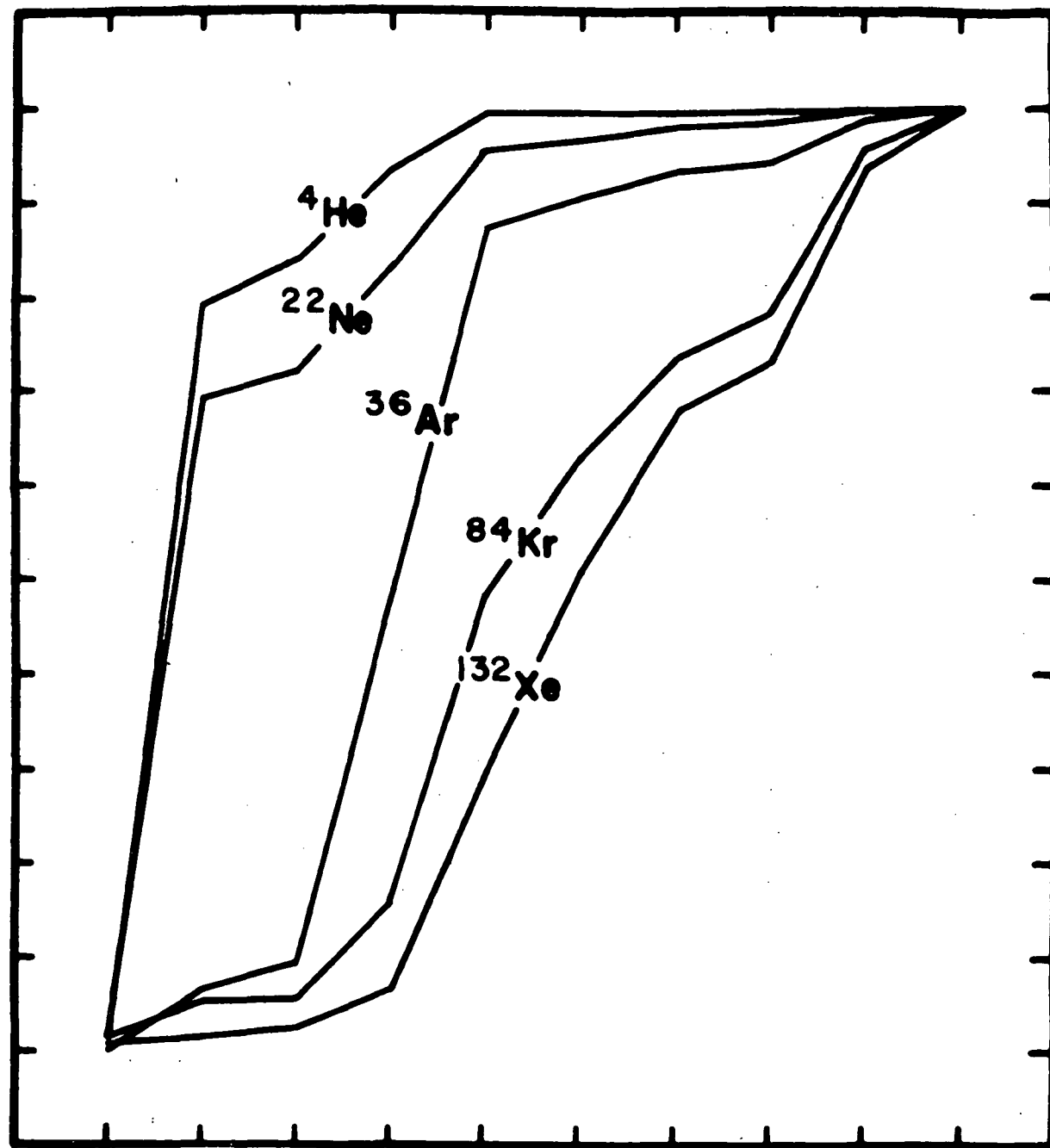


Figure 6. The release pattern of noble gases from 15601.64 at extraction temperatures $\geq 600^{\circ}\text{C}$. The preferential release of light weight noble gases at low extraction temperatures is paralleled by isotopic fractionation across the isotopes of each noble gas.

CUMULATIVE PERCENT RELEASED →

100
90
80
70
60
50
40
30
20
10
0



600 800 1000 1200 1400

EXTRACTION TEMPERATURE →

Figure 7. Correlation of the xenon isotopes released from sample 15601.64. The solid lines passing through atmospheric xenon are defined by eq. (1). The broken lines through AVCC $^{134}\text{Xe}/^{132}\text{Xe}$ and $^{136}\text{Xe}/^{132}\text{Xe}$ ratios also fit isotopic fractionation, and the broken line labeled 129' would pass through atmospheric $^{129}\text{Xe}/^{132}\text{Xe}$ which had been corrected for radiogenic ^{129}Xe in air (BOULOS and MANUEL, 1971). The ^{126}Xe isotope is shown as darkened points. This isotope shows the largest scatter from the fractionation line, due to the large effect of spallation products on the $^{126}\text{Xe}/^{132}\text{Xe}$ ratio..

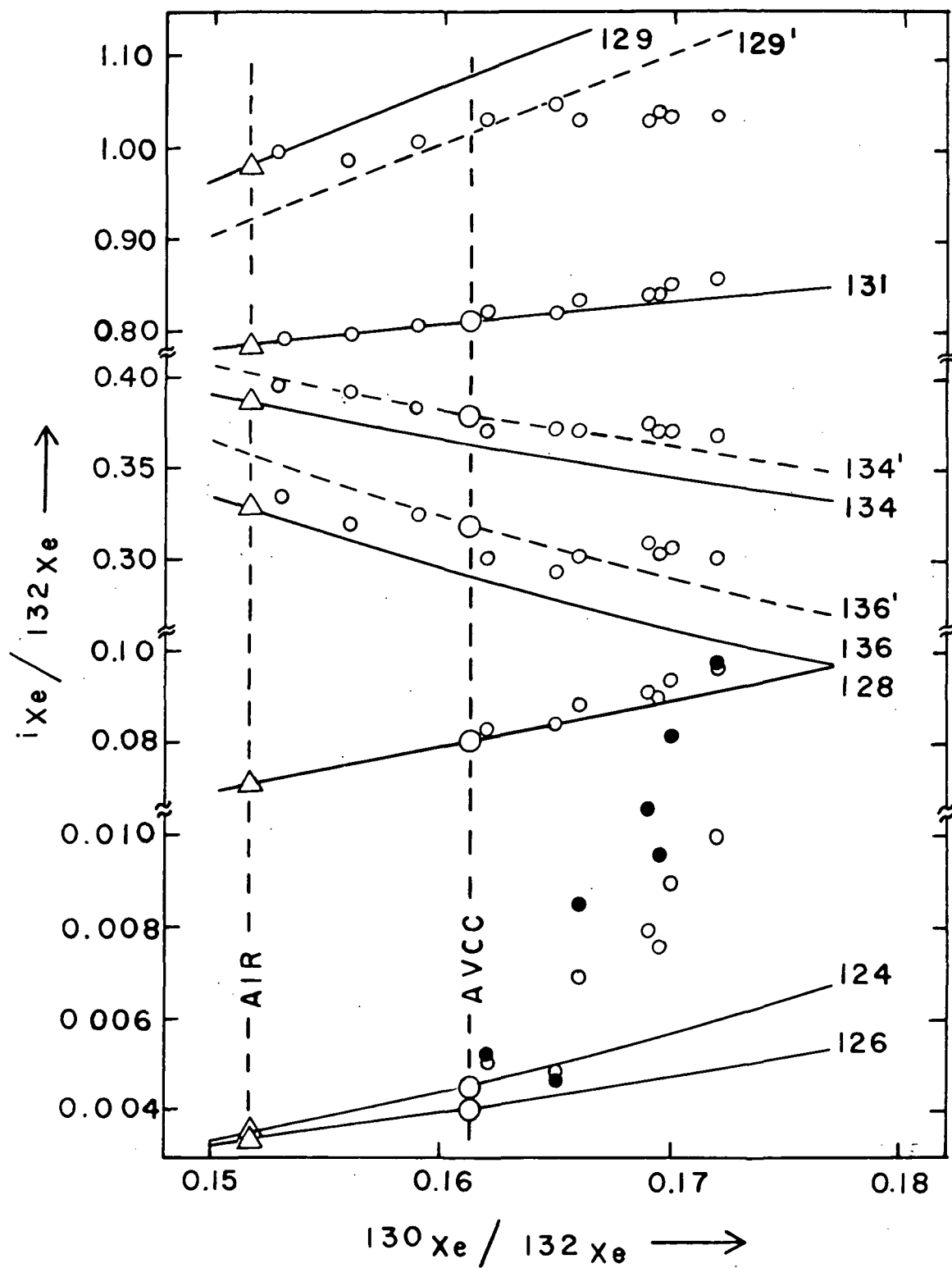
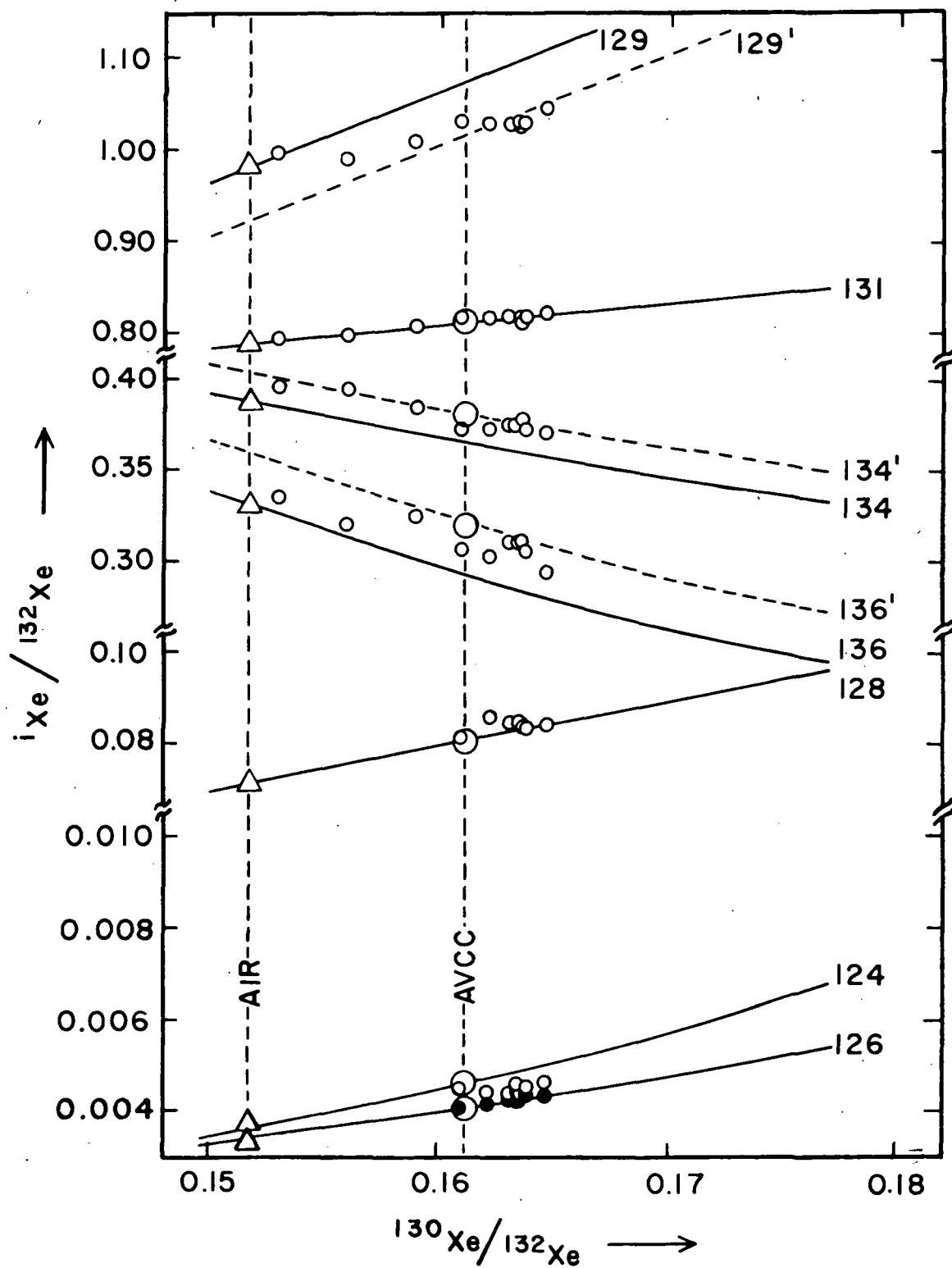


Figure 8. Correlation of the spallation-free xenon isotopes from sample 15601.64. The heavy xenon isotopes are enriched above the atmospheric fractionation line, as are the heavy xenon isotopes in AVCC xenon. The ^{124}Xe isotope appears to be depleted relative to fractionated atmospheric xenon.



APPENDIX II

On the Isotopic Composition of Noble Gases in Lunar
Fines (15271, 65), in Meteorites and in the Atmosphere

Figures and Tables from a manuscript
in preparation for submission to
Zeitschrift für Naturforschung

Table 1. Helium, neon and argon from stepwise heating of lunar fines 15271, 65

Temp. (°C)	Isotopic Ratios					Gas Content ($\times 10^{-8}$ cc STP/g)		
	$^4\text{He}/^3\text{He}$	$^{20}\text{Ne}/^{22}\text{Ne}$	$^{21}\text{Ne}/^{22}\text{Ne}$	$^{38}\text{Ar}/^{36}\text{Ar}$	$^{40}\text{Ar}/^{36}\text{Ar}$	^4He	^{22}Ne	^{36}Ar
600°	1963 ± 12	13.78 ± 0.03	0.0337 ± 0.0001	0.188 ± 0.001	31.2 ± 0.1	6.41	53.2	5.45
800°	2463 ± 4	13.12 ± 0.03	0.0342 ± 0.0001	0.179 ± 0.001	2.90 ± 0.01	3,820,000	20,300	34,300
1000°	2722 ± 8	12.79 ± 0.02	0.0359 ± 0.0001	0.185 ± 0.004	0.805 ± 0.003	83,000	103,000	184,000
1200°	2654 ± 5	12.84 ± 0.04	0.0396 ± 0.0001	0.189 ± 0.001	1.03 ± 0.00	67,400	6,710	22,800
1400°	2450 ± 7	12.20 ± 0.01	0.0421 ± 0.0001	0.188 ± 0.001	1.23 ± 0.00	169	1,780	27,400
1600°				0.228 ± 0.001	3.77 ± 0.01	72.8		21.8
AIR	7.7×10^5	9.81	0.0290	0.187	296			

Table 2. Krypton released from stepwise heating of lunar fines 15271, 65

Temp. (°C)	^{86}Kr Released $\times 10^{-10}$ cc STP/g	^{78}Kr	^{80}Kr	^{82}Kr	^{83}Kr	^{84}Kr	^{86}Kr
800°C	22.7	2.46 ± 0.02	14.7 ± 0.1	70.9 ± 0.3	69.9 ± 0.3	335 ± 1	≤ 100
1000°C	420	2.14 ± 0.02	13.4 ± 0.1	68.4 ± 0.3	67.4 ± 0.3	329 ± 1	≤ 100
1200°C	730	2.26 ± 0.02	13.7 ± 0.1	67.7 ± 0.2	67.8 ± 0.2	328 ± 1	≤ 100
1400°C	159	2.66 ± 0.01	14.7 ± 0.1	69.2 ± 0.1	70.0 ± 0.1	329 ± 2	≤ 100
1600°C	1.15	3.89 ± 0.02	17.7 ± 0.1	74.0 ± 0.2	76.4 ± 0.2	333 ± 1	≤ 100
AIR		2.02	13.0	66.2	66.0	327	≤ 100
AVCC		1.92	12.6	64.9	65.1	323	≤ 100

Table 3. Xenon released from stepwise heating of lunar fines 15271, 65

Temp. (°C)	^{132}Xe cc STP/g $\times 10^{-10}$	^{124}Xe	^{126}Xe	^{128}Xe	^{129}Xe	^{130}Xe	^{131}Xe	^{132}Xe	^{134}Xe	^{136}Xe
600°	0.15			7.25 ± 0.14	97.8 ± 0.7	15.0 ± 0.1	78.0 ± 0.6	≤ 100	38.7 ± 0.2	33.5 ± 0.2
800°	1.5	0.513 ± 0.005	0.569 ± 0.008	7.98 ± 0.04	100.1 ± 0.5	15.7 ± 0.1	80.4 ± 0.4	≤ 100	38.3 ± 0.2	32.1 ± 0.1
1000°	37	0.547 ± 0.002	0.566 ± 0.003	8.47 ± 0.04	104.9 ± 0.5	16.5 ± 0.1	83.1 ± 0.4	≤ 100	36.4 ± 0.2	29.4 ± 0.1
1200°	130	0.770 ± 0.006	0.974 ± 0.007	8.99 ± 0.06	104.6 ± 0.6	16.8 ± 0.1	84.1 ± 0.5	≤ 100	36.5 ± 0.2	29.4 ± 0.2
1400°	66	1.10 ± 0.00	1.54 ± 0.00	9.81 ± 0.03	104.8 ± 0.4	17.4 ± 0.1	86.1 ± 0.5	≤ 100	36.8 ± 0.1	29.8 ± 0.1
1600°	1.0	1.27 ± 0.00	1.87 ± 0.01	10.37 ± 0.06	106.2 ± 0.4	17.8 ± 0.1	89.0 ± 0.4	≤ 100	36.8 ± 0.1	29.9 ± 0.1
AIR		0.357	0.335	7.14	98.3	15.2	78.8	≤ 100	38.8	33.0
AVCC		0.452	0.406	8.09		16.1	81.5	≤ 100	38.1	32.0

Figure 1

Neon isotopes released by stepwise heating of lunar fines (15271, 65). The triangle (Δ) represents atmospheric neon and the line passing through atmospheric neon shows the effects of mass fractionation. The approximately vertical lines extrapolate to cosmogenic neon.

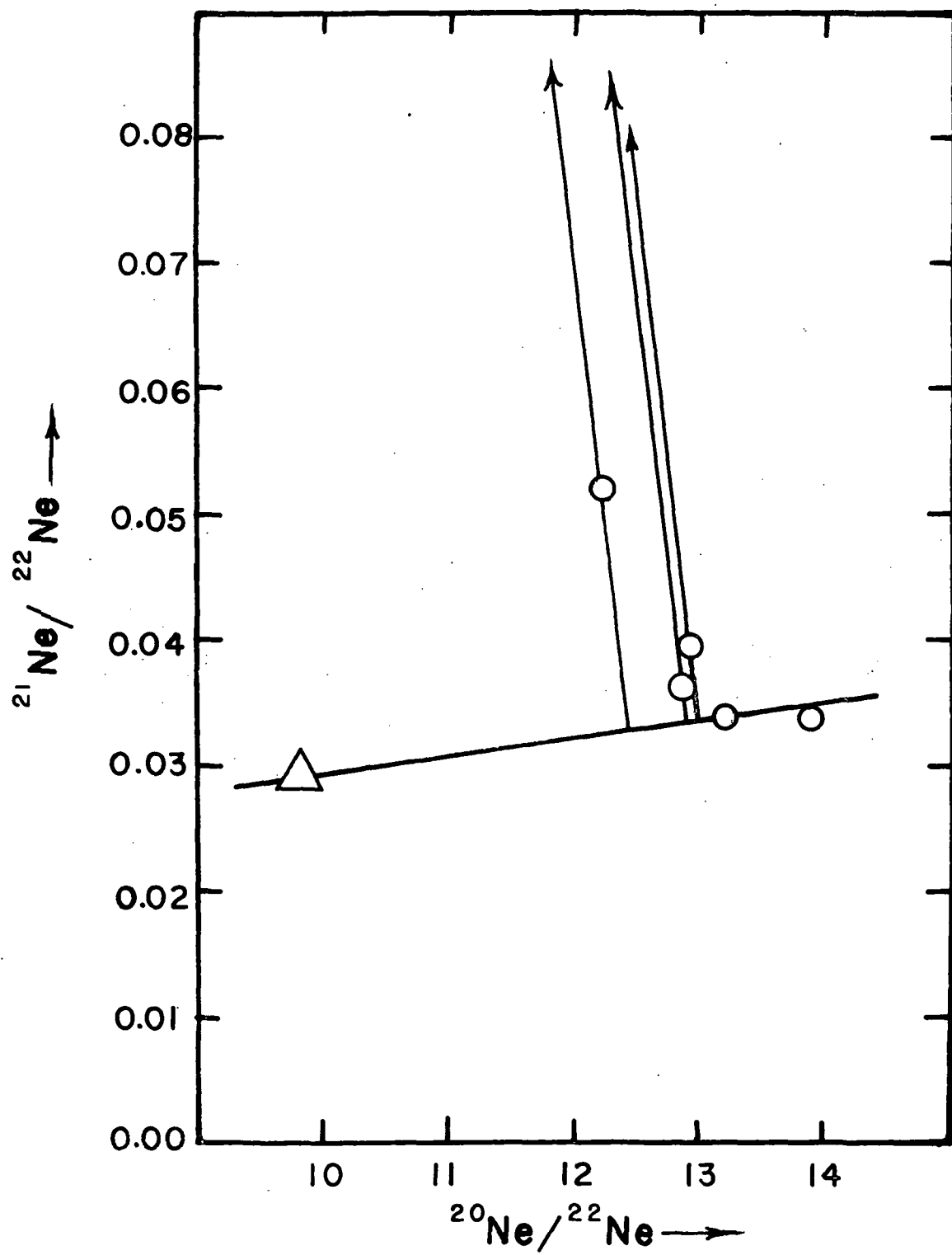


Figure 2

The release pattern of noble gases from lunar fines (15271, 65).

The preferential release of light-weight noble gases at low extraction temperatures parallels the behavior observed for the neon isotopes.

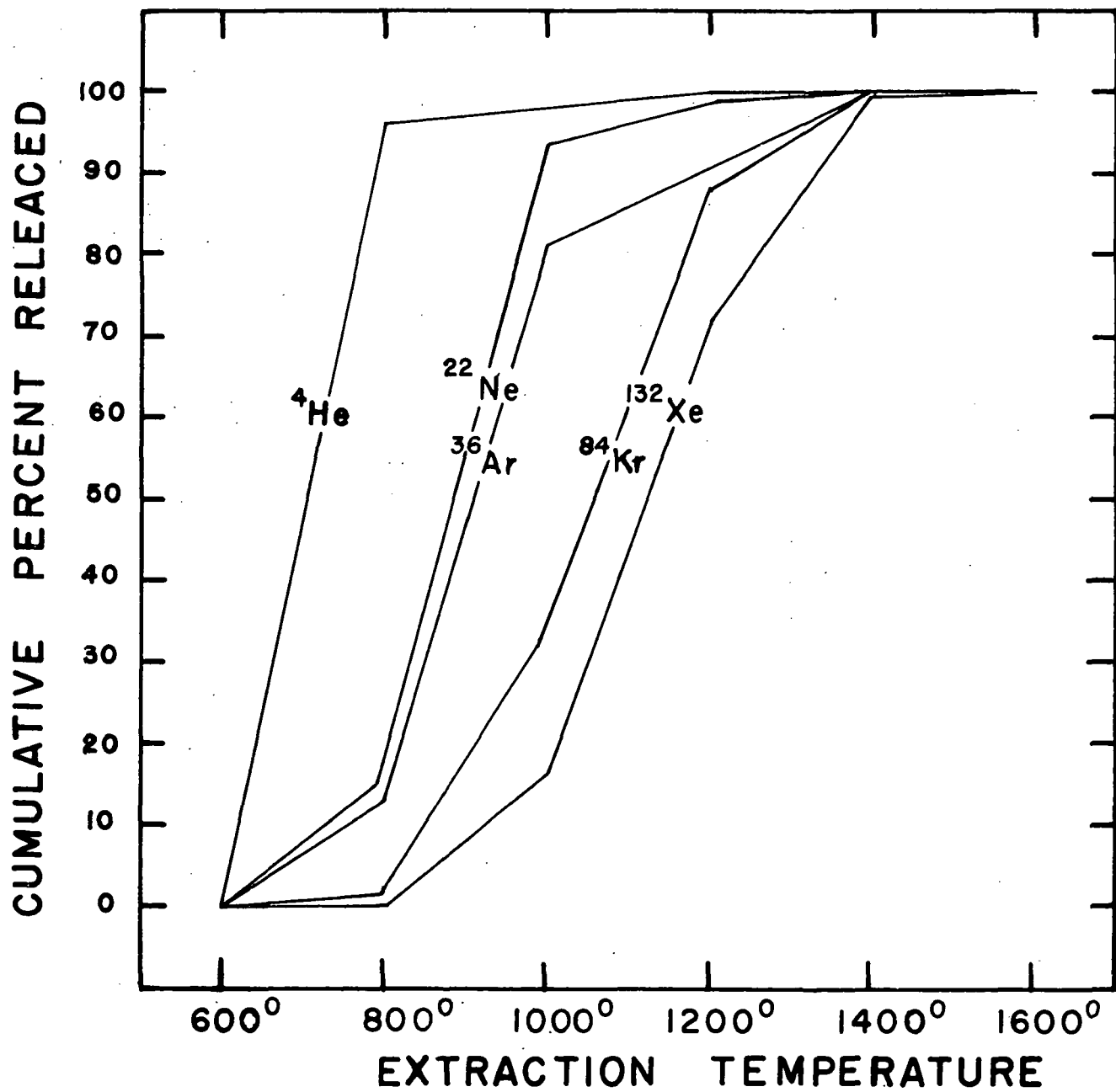


Figure 3

Krypton isotopes released by stepwise heating of lunar fines (15271, 65). The lines labeled 83, 82, 80 and 78 show the effect of isotopic fractionation predicted by Eq. (1). The krypton released at 1600°C is shown by solid circles (●) and the lines passing through these points extrapolate to pure cosmogenic krypton.

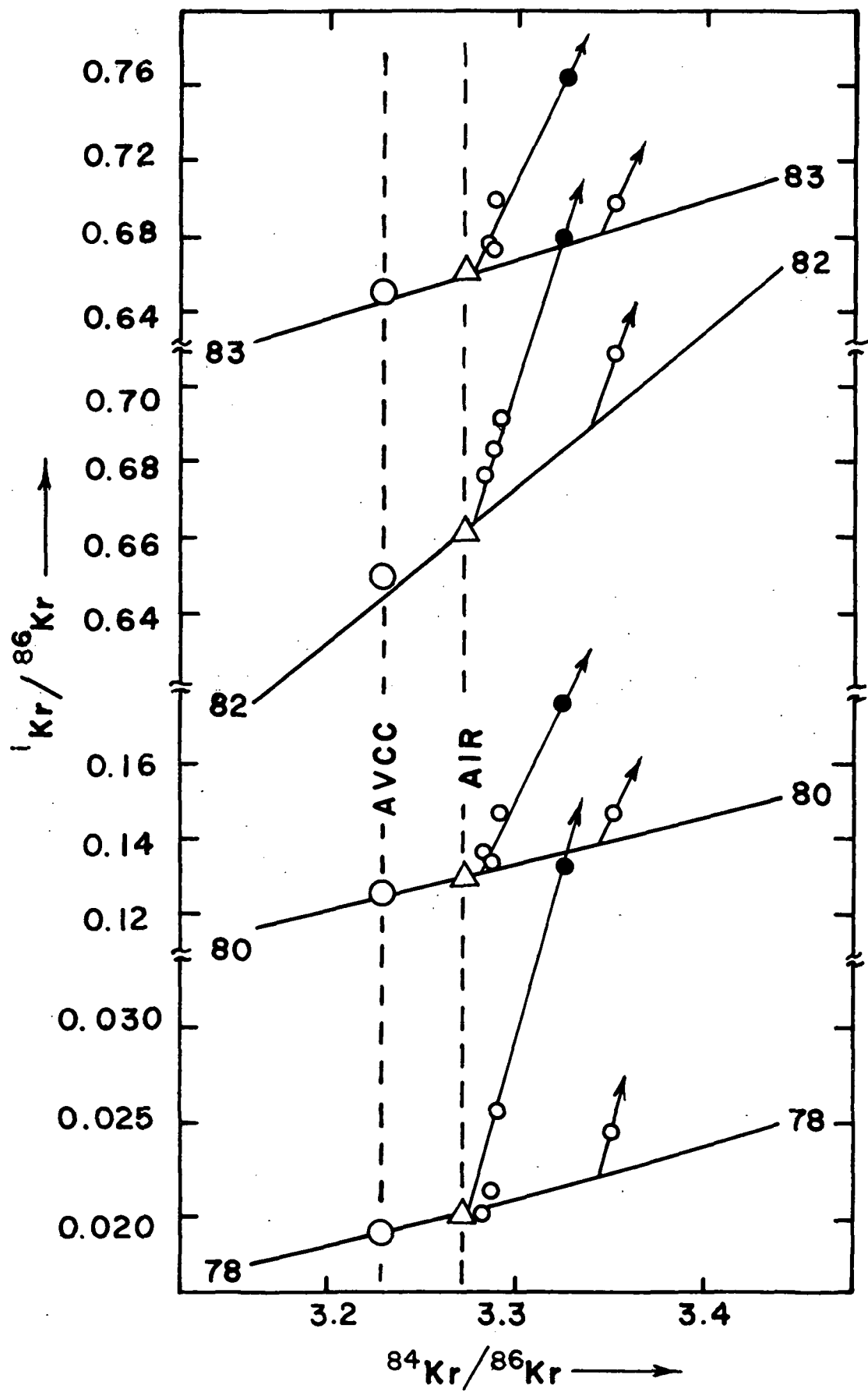


Figure 4

Xenon isotopes released by stepwise heating of lunar fines (15271, 65). The $^{126}\text{Xe}/^{132}\text{Xe}$ ratios are represented by dark circles (●). The lines labeled 129, 131, 134, 136, 128, 124 and 126 show the effects of fractionation on atmospheric xenon. The lines labeled 134' and 136' show the effects of fractionation on AVCC-type xenon, and the line labeled 129' represents fractionation of atmospheric xenon which has been corrected for the radiogenic ^{129}Xe .

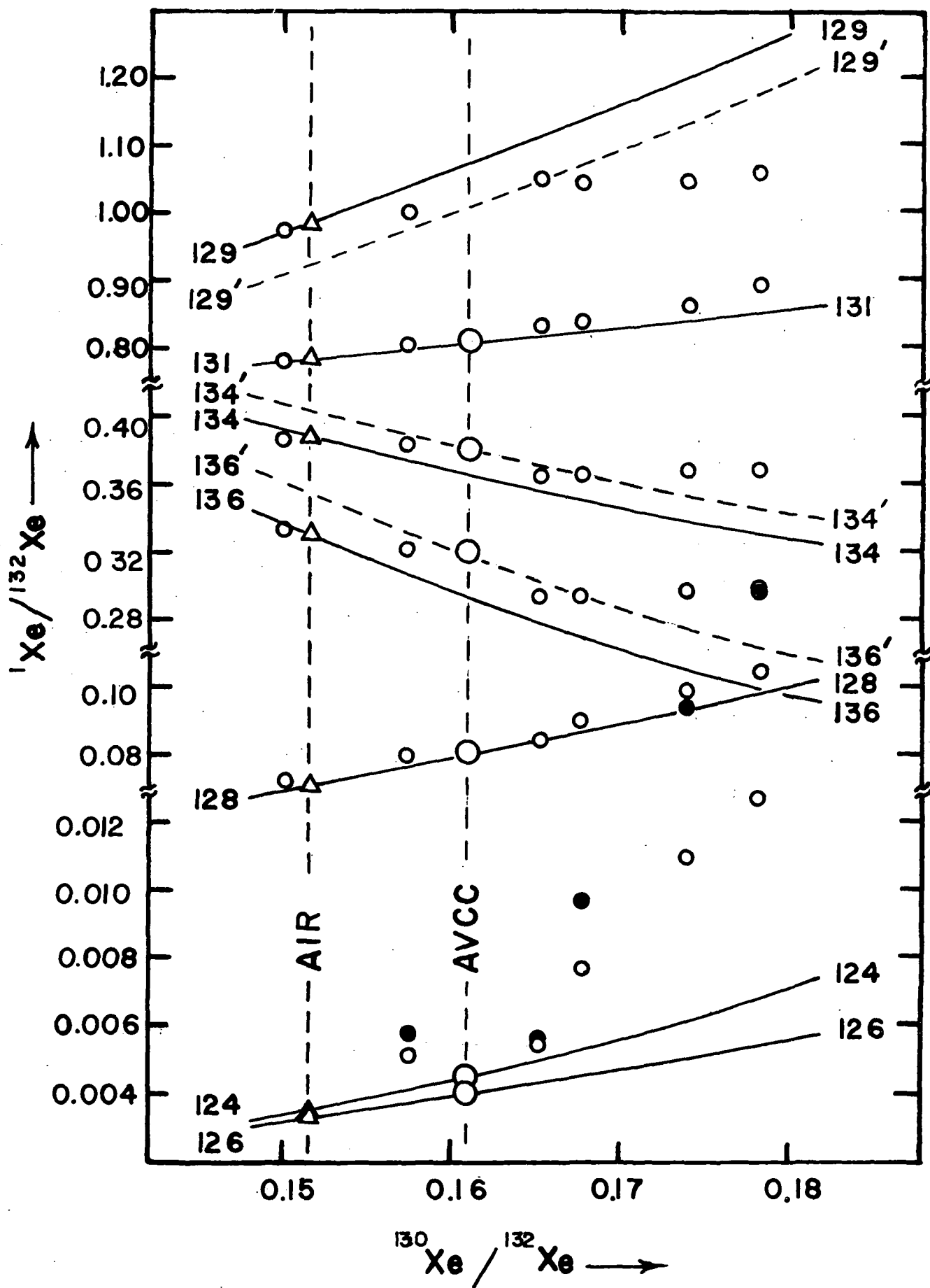
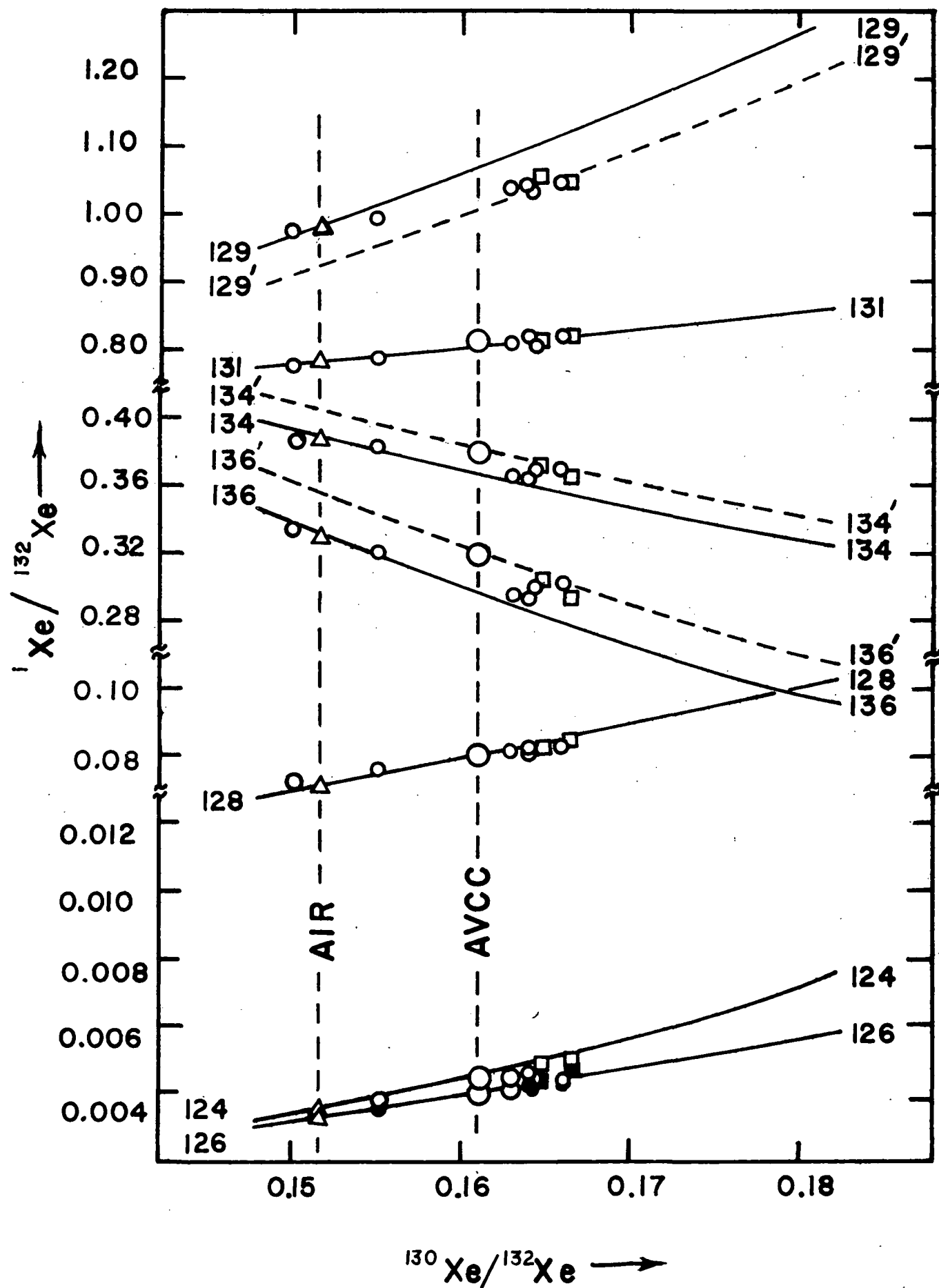


Figure 5

Xenon isotopes from lunar fines (15271, 65) after subtracting spallation components. The corrected isotope ratios fit closely to the pattern predicted by Eq. (1) for fractionated atmospheric xenon, except for an excess of ^{136}Xe and ^{134}Xe .



APPENDIX III

Xenon in Carbonaceous Chondrites

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Xenon in Carbonaceous Chondrites

O. K. Manuel and E. W. Hennecke
Nuclear Division
Department of Chemistry
University of Missouri
Rolla, Missouri 65401, U.S.A.

and

D. D. Sabu
Department of Chemistry
Grambling College
Grambling, Louisiana 71254, U.S.A.

ABSTRACT

Carbonaceous chondrites contain two isotopically distinct components of trapped xenon which cannot be explained by the occurrence of nuclear or fractionation processes within these meteorites.

Several authors¹⁻⁷ have noted an enrichment of ^{134}Xe and ^{136}Xe in the xenon released from carbonaceous chondrites at extraction temperatures of about $800^{\circ}\text{--}1000^{\circ}\text{C}$. Reynolds and Turner¹ first observed this isotopic anomaly of xenon in a stepwise heating experiment on the carbonaceous chondrite, Renazzo. They suggested that the isotopic anomaly of xenon released at $800^{\circ}\text{--}1000^{\circ}$ might be due to the spontaneous fission of ^{244}Pu but further noted that the presence of small amounts of ^{124}Xe and ^{126}Xe would have to be explained by some additional process. Subsequent studies of xenon in other carbonaceous chondrites have confirmed the existence of a "Renazzo type" isotopic anomaly. However, little attention has been focused on the light xenon isotopes and most hypotheses on the origin of the "Renazzo-type" anomaly have been primarily concerned with the possible enrichment of heavy xenon isotopes from neutron induced fission², from a 'carrier' of heavy xenon adsorbed on carbonaceous material⁵, from fission of superheavy elements⁸⁻¹¹ or from mass fractionation¹².

To demonstrate that the "Renazzo-type" anomaly includes both the lightest and heaviest isotopes of xenon, we present in Table 1 the isotopic data for carbonaceous chondrites at those extraction temperatures which show the most spectacular enrichments of the heavy xenon isotopes. The isotopic composition of xenon in air¹³ and that of the total trapped xenon in average carbonaceous chondrites^{14, 15} (AVCC) are shown for comparison. The bulk of the trapped xenon is released from each of the chondrites at higher extraction temperatures and consists of an isotopic spectrum similar to AVCC xenon.

The xenon from carbonaceous chondrites is shown in Table 1 in the order of increasing enrichments of the heavy xenon isotopes. This arrangement of the xenon data also corresponds to an increasing enrichment of the lighter xenon isotopes; a correlation which cannot be accounted for by the addition of fission products^{1-5, 8-11}. Further, this correlation is opposite to that expected from mass dependent fractionation^{6, 7, 12}. Spallation produced xenon isotopes are most easily observed at ^{124}Xe and ^{126}Xe , where all known spallation yields^{16, 17} cause the $^{124}\text{Xe}/^{126}\text{Xe}$ ratio to decrease below that of AVCC xenon. The absence of any significant spallation contribution to the light xenon isotopes in the "Renazzo type" anomaly is indicated by the fact that all of the $^{124}\text{Xe}/^{126}\text{Xe}$ ratios are higher than that in AVCC xenon and further these $^{124}\text{Xe}/^{126}\text{Xe}$ ratios increase with the $^{136}\text{Xe}/^{130}\text{Xe}$ ratios.

Fig. 1 shows the observed correlation of the $^{124}\text{Xe}/^{130}\text{Xe}$ ratios with the $^{136}\text{Xe}/^{130}\text{Xe}$ ratios in these carbonaceous chondrites and the correlations expected from mixtures of AVCC xenon with fission products or with spallation products. The pattern expected from mass dependent fractionation is also shown. It is obvious from the data in Table 1 and Fig. 1 that the observed correlation cannot be accounted for by any of the previously proposed mechanisms. We know of no plausible in situ nuclear process which could produce both an enrichment in the lightest and the heaviest isotopes of xenon. Further, the high degree of correlation of $^{124}\text{Xe}/^{130}\text{Xe}$ with $^{136}\text{Xe}/^{130}\text{Xe}$ (98%) seems to rule out any accidental mixture of in situ produced enrichments of the heaviest and the lightest isotopes. We therefore

conclude that the "Renazzo type" anomaly is due to an isotopically distinct component of xenon (hereafter referred to as X) which has been incorporated into the carbonaceous chondrites. The correlations for the xenon isotope ratios seen in Table 1 and Fig. 1 are then the result of mixtures of AVCC xenon with this component X.

Recently it has been suggested that atmospheric and solar-wind implanted xenon are related by isotopic mass fractionation^{18,19}. The dashed line through atmospheric xenon in Fig. 1 shows the effect expected from Aston's equation²⁰ for diffusive fractionation. Also shown is a recent estimate of the isotopic composition of solar-wind implanted xenon obtained from analysis of lunar soils¹⁹. The shift of AVCC xenon from the fractionation line relating atmospheric and solar xenon suggests that AVCC xenon may contain a small component of X-type xenon.

The xenon released from Allende at 800°C contains a higher percentage of X-type xenon than does any other xenon reported to date. We therefore use this xenon to seek information on the relative abundances of the isotopes and possible origin of component X. From the data shown in Table 1 it is obvious that X-type xenon shows the greatest enrichment in the lighter isotopes, $^{124}, ^{126}\text{Xe}$, and in the heavier isotopes, $^{134}, ^{136}\text{Xe}$, relative to ^{130}Xe . These are the isotopes of xenon expected to be produced in supernova explosions²¹, the $^{124}, ^{126}\text{Xe}$ isotopes by the p-process and the $^{134}, ^{136}\text{Xe}$ by the r-process. Thus, it is possible that the X-type xenon in carbonaceous chondrites represents material that has been added to our solar system from a nearby supernova. However, no evidence for the

addition of products from a separate nucleosynthesis event has been found in the other elements.

In order to look for a more quantitative relationship between X-type xenon and that in the earth's atmosphere, we employ the method of Canalas et al.²² to compute a fractionation factor, F^m , for each xenon isotope.

$$F^m = (^{i}\text{Xe}/^{130}\text{Xe})_{\text{Met}} / (^{i}\text{Xe}/^{130}\text{Xe})_{\text{Air}} \quad (1)$$

Fig. 2 shows the values of F^m for the xenon isotopes released from Allende at 800°C. The values of F^m for the light isotopes, $^{124}\text{-}^{130}\text{Xe}$, decrease in a regular manner with increasing mass number, but the F^m values for the heavier isotopes, $^{131}\text{-}^{136}\text{Xe}$, show an irregular pattern with increasing mass number. The pattern seen across the lighter xenon isotopes is suggestive of mass dependent fractionation, a process which has recently been recognized as the source for many isotopic anomalies of noble gases^{6, 7, 12, 23-25}. The dashed line in Fig. 2 shows the pattern expected across the fractionation factors, F^m , if atmosphere xenon and X-type xenon are related by the diffusive gas loss equation of Aston^{20, 25}. The dashed line corresponds to a volume change in xenon of $\approx 3 \times 10^7$. The abundance pattern of the lighter isotopes, $^{124}\text{-}^{130}\text{Xe}$, fit Aston's equation and there remains an excess of the heavier xenon isotopes in the ratio $^{131}\text{Xe} : ^{132}\text{Xe} : ^{134}\text{Xe} : ^{136}\text{Xe} = 0.30 \pm 0.11 : 0.58 \pm 0.14 : 1.00 : 1.41 \pm 0.10$. Thus, the correlation shown in Fig. 2 suggests that the X-type xenon which has been incorporated into carbonaceous chondrites can be related to atmospheric xenon by a combination of isotopic mass fractionation plus some process which produced the heavy xenon isotopes in the above ratio.

In considering possible mechanisms for the production of excess $^{131-136}\text{Xe}$ it should be kept in mind that the correlations shown in Table 1 and Fig. 1 indicate that the isotopic anomalies of the X-type xenon were not produced in situ but were present when this xenon was incorporated into the carbonaceous chondrites.

Srinivasan et al.⁸ first suggested the possibility that the enrichment of heavy xenon isotopes in carbonaceous chondrites might be due to the spontaneous fission of superheavy elements. Recognition that the enrichment of these heavy xenon isotopes was not produced within the meteorite makes this hypothesis more attractive, since the proposed superheavy elements need not survive the time interval ($\approx 10^8$ years) between the end of nucleosynthesis and the formation of the meteorites²⁶. However, in the absence of quantitative information on the fission yields of superheavy elements or their synthesis in the r-process, this hypothesis must be considered speculative. No known spontaneous fission yields fit the abundance pattern of excess $^{131-136}\text{Xe}$ in X-type xenon.

Pepin² found an enrichment pattern of the heavy xenon isotopes in the Renazzo, Murray and Orgueil carbonaceous chondrites similar to that shown above in X-type xenon. He suggested that the relative abundances of the heavy xenon isotopes could have resulted from an extremely high flux of thermal neutrons on ^{235}U . However, Pepin² failed to consider the abundances of the lighter isotopes, $^{124,126}\text{Xe}$, and concluded that the absence of detectable neutron-capture anomalies in the rare earths of meteorites left no satisfactory explanation for the excess heavy xenon isotopes.

Recognition of the correlation between this isotopic anomaly of heavy xenon isotopes with an isotopic anomaly of the light xenon isotopes (Fig. 1) makes in situ production of these anomalies unnecessary. Thus, neutron induced fission of ^{235}U becomes a feasible mechanism for the production of excess $^{131-136}\text{Xe}$ in the X-type xenon. Due to the large thermal neutron capture cross-section of ^{135}Xe , a thermal flux of $\approx 2.8 \times 10^{13}$ neutrons $\text{cm}^{-2} \text{ sec}^{-1}$ on ^{235}U would produce $^{131}\text{Xe}:^{132}\text{Xe}:^{134}\text{Xe}:^{136}\text{Xe} = 0.36:0.54:1.00:1.41$. Within the statistical error limits ($\pm \sigma$) these yields agree with those calculated above in X-type xenon. The conditions which prevailed during the irradiation may resemble those in the model proposed by Fowler et al.²⁷ for the solar synthesis of D, Li, Be and B. In this case the X-type xenon might represent a sample from the small fraction of solar material irradiated with thermal neutrons.

A third possible mechanism for the production of excess $^{131-136}\text{Xe}$ is that of mass dependent fractionation. It has been shown earlier that changes in the relative abundances of the heavy xenon isotopes, $^{131-136}\text{Xe}$, released from carbonaceous chondrites follows closely to the pattern expected from isotopic fractionation^{6, 7, 12, 25}. However when Manuel et al.⁷ considered correlations for all the xenon isotopes released from Allende and Mokoia, they noted that the abundances of ^{124}Xe and ^{126}Xe were too high in those temperature fractions showing excess ^{136}Xe to be accounted for by a single fractionating process. In view of the correlation of excess ^{124}Xe with excess ^{136}Xe (Fig. 1) the explanation of both by isotopic fractionation would require that X-type xenon consist of a mixture of two xenon

components severely fractionated in opposite directions relative to atmospheric xenon. Although such a mixture of fractionated components seems unlikely, this possibility should not be ruled out until we have a better understanding of the physical environments capable of producing X-type xenon from other sources.

In summary, the enrichment of heavy xenon isotopes released from carbonaceous chondrites near 800°-1000°C is accompanied by an enrichment of the light xenon isotopes. The high degree of correlation between these two isotopic anomalies suggests that both result from a common source. Since no known nuclear or physical processes are capable of producing both anomalies in situ, it is suggested that they result from the release of isotopically anomalous xenon (component X) that was trapped in the meteorites. The light xenon isotopes, $^{124-130}\text{Xe}$, in this X-type xenon can be related to atmospheric or solar-type xenon by mass fractionation. The astronomical setting which produced this and the excess heavy xenon isotopes in X-type xenon is not well understood, but recognition that this anomalous xenon was trapped in the meteorites removes many of the limitations on hypotheses previously proposed for the in situ production of heavy xenon isotopes. Additional information on the solar distribution of X-type xenon may be obtained from isotopic analyses of xenon implanted in lunar soil by the solar wind.

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Table 1 Xenon Isotopes in Carbonaceous Chondrites

Sample	124	126	128	130	131	132	134	136	References
AIR	2.353	2.206	47.03	100	519.1	659.1	255.9	217.4	Nier ¹³
AVCC	2.828	2.533	50.38	100	506.7	620.9	236.9	199.0	Marti ¹⁴ and Eugster et al. ¹⁵
Renazzo 800°	2.93 ± 0.05	2.57 ± 0.03	50.6 ± 0.4	100	505.8 ± 2.4	617.2 ± 1.9	243.3 ± 2.2	210.4 ± 1.5	Reynolds and Turner ¹
Leoville 1010°	3.07 ± 0.09	2.66 ± 0.12	51.4 ± 0.5	100	511.3 ± 4.9	625.0 ± 3.9	250.0 ± 2.4	220.6 ± 2.3	Manuel et al. ⁶
Makoia (Matrix) 600°	3.15 ± 0.09	2.72 ± 0.09	52.7 ± 0.7	100	519.9 ± 6.8	633.3 ± 7.6	276.1 ± 3.6	248.9 ± 3.6	Rowe ⁵
Makoia 750°	3.39 ± 0.06	2.90 ± 0.05	52.0 ± 0.8	100	514.1 ± 5.6	624.5 ± 5.6	278.6 ± 3.2	261.1 ± 2.0	Manuel et al. ⁷
Allende 1000°	3.39 ± 0.14	2.77 ± 0.09	54.5 ± 0.9	100	513.0 ± 6.1	624.9 ± 8.6	291.2 ± 3.9	282.5 ± 2.4	Manuel et al. ⁷
Allende 800°	3.62 ± 0.13	2.84 ± 0.09	52.1 ± 1.5	100	518.6 ± 11.9	641.0 ± 14.3	305.1 ± 7.0	299.4 ± 5.4	Manuel et al. ⁷

Fig. 1. The correlation observed between the enrichment of the $^{124}\text{Xe}/^{130}\text{Xe}$ ratios and the $^{136}\text{Xe}/^{130}\text{Xe}$ ratios in carbonaceous chondrites which exhibit the "Renazzo type" anomaly. The isotope data for air, AVCC and solar xenon are from Nier¹³, Marti¹⁴ and Eugster et al.¹⁵, and Kaiser¹⁹, respectively. and the isotope data from stepwise heating of carbonaceous chondrites are from Reynolds and Turner¹, Manuel et al.⁶, Rowe⁵ and Manuel et al.⁷.

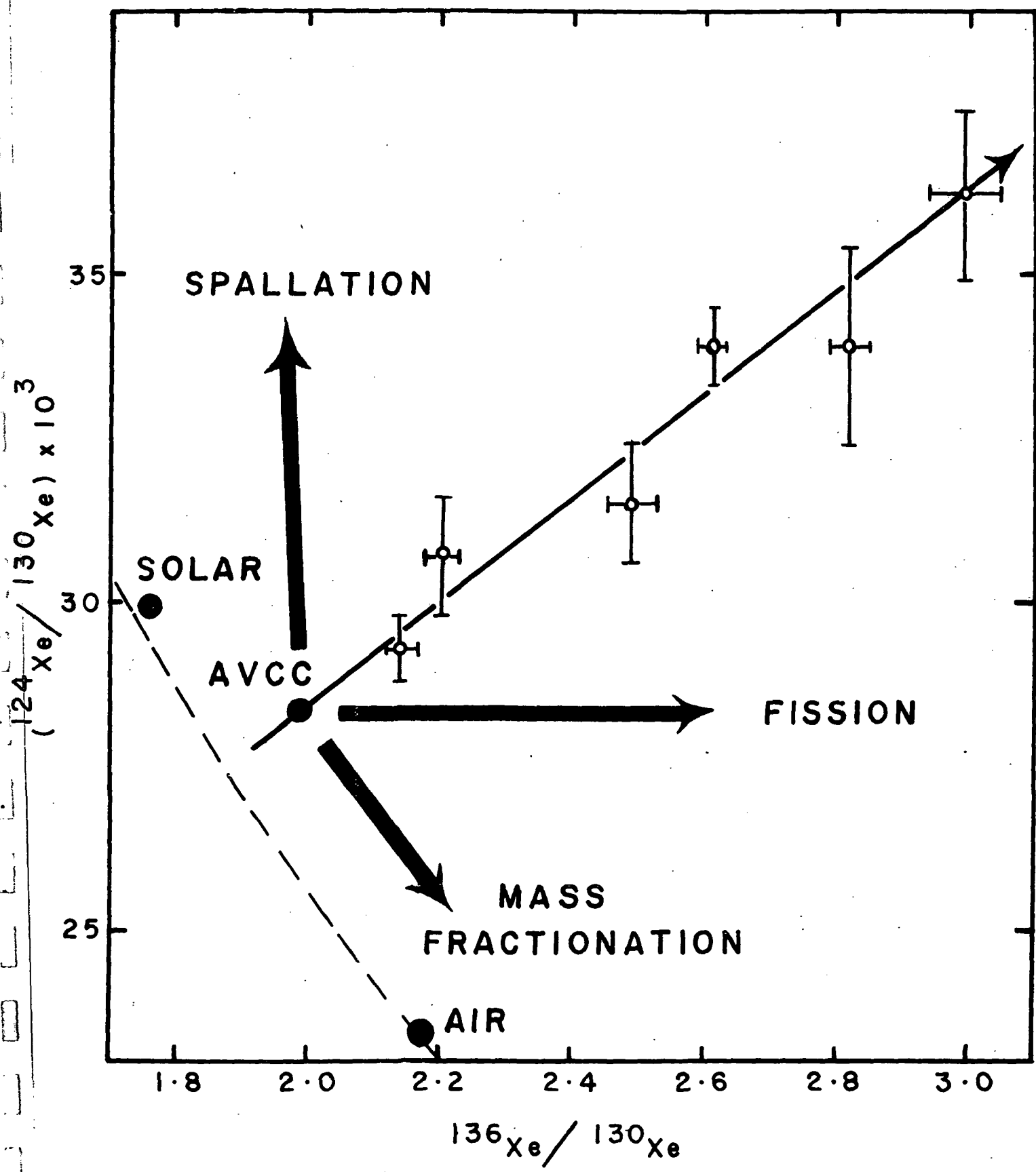


Fig. 2. A comparison of the abundance pattern of xenon isotopes from the 800° fraction of Allende⁷ relative to the abundance of xenon isotopes in the atmosphere¹³. The dashed line shows the mass fractionation effects calculated from Aston's equation²⁰ for diffusive gas loss.

